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Variability and Source Apportionment of Ultrafine Particles in Urban Europe

Meritxell Garcia Marlès

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PhD Thesis

VARIABILITY AND SOURCE APPORTIONMENT OF ULTRAFINE PARTICLES IN URBAN EUROPE

Meritxell Garcia Marlès



UNIVERSITAT DE
BARCELONA

VARIABILITY AND SOURCE APPORTIONMENT OF ULTRAFINE PARTICLES IN URBAN EUROPE

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“The little things are infinitely the most important.”

Arthur Conan Doyle

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ABSTRACT

Atmospheric particulate matter (PM) is recognised as a major air pollutant with significant impacts on human health and the Earth's climate. The nature and severity of its effects depend, among other factors, on particle size. Ultrafine particles (UFPs), defined as particles with diameters below 100 nm, contribute only a small fraction of PM mass concentrations ($\mu\text{g m}^{-3}$) but represent a substantial portion of particle number concentrations (PNCs, $\# \text{ cm}^{-3}$). Particle number size distributions (PNSDs) categorise particle into three size modes: Nucleation ($< 25 \text{ nm}$), Aitken (25–100 nm) and accumulation (100–1000 nm).

Due to their nanoscale size, UFPs can penetrate deep into the respiratory tract and translocate through the alveolar barrier into the bloodstream, reaching other organs. However, evidence on their health impacts remain inconsistent, likely due to the scarcity of long-term UFP datasets, the lack of standardised monitoring methods, the strong spatial variability of UFPs, and the diversity of sources contributing to PNCs in different regions. Currently, UFPs have no legally binding standards at the EU or global levels. The new Ambient Air Quality Directive (European Union) 2024/2881 introduced UFPs into European legislation for the first time. Although binding limits are not yet defined, it mandates monitoring of PNC at urban supersites, rural supersites, and pollution hotspots such as traffic-dense areas, airports, harbours, and industrial locations, while PNSD monitoring is required only at urban supersites, covering particles down to 10 nm. The goal is to obtain harmonised data to subsequently evaluate health associations, thereby supporting decisions on evidence-based air quality (AQ) management.

The objective of this thesis is to improve understanding of the spatial and temporal variability and sources of UFPs in urban Europe. Long-term PNC and PNSD data (2009–2019) from twenty-nine European cities were compiled, harmonised, and analysed. Spatial variability and long-term trends of UFPs across diverse environments were investigated, and sources were identified and quantified through source apportionment (SA) analyses using Positive Matrix Factorisation (PMF). In addition, aviation-related emissions near a major European airport were characterised, and meteorology-driven dispersion effects on source contributions were assessed.

The results presented provide one of the most comprehensive assessments of UFP concentrations and SA across European urban environments to date. Harmonisation of PNSDs within the 10–800 nm size range allowed reliable inter-site comparisons and minimised artefacts arising from different lower size detection limits. PNCs increased progressively from Northern to Southern Europe, with substantially higher levels at traffic sites, pointing to road traffic as the dominant contributor to UFPs in cities. Long-term trend analyses (2009–2019) revealed marked decreases in Aitken and accumulation mode particles, driven by reductions in traffic-related emissions following successive European road traffic emission policies. In contrast, nucleation mode particles remained stable or increased slightly, influenced by new particle formation (NPF) events and non-traffic sources such as industry, shipping, and aviation.

SA using PMF identified up to ten distinct factors contributing to PNCs, with road traffic as the major contributor, followed by photochemical nucleation and domestic heating. The source contributions varied a lot among cities, in both type and magnitude. Surprisingly, NPF contributions were not the highest in Southern Europe (with higher insolation and biogenic emissions of precursors) but in Central Europe, likely due to SO₂ emissions from coal-fired power plants. Domestic heating was a major source of UFPs in a relevant number of cities.

Aviation-related emissions were characterised separately in a dedicated case study at Brussels Airport, where aircraft operations were found to dominate local UFP levels and remained detectable several kilometres downwind. The application of dispersion normalisation in a Barcelona analysis (2013–2024) further improved SA results by separating emission-driven changes from meteorological variability. The 2013–2024 UFP decreasing trend was more pronounced than for 2013–2019, reflecting the effect of European, national, regional, and local measures on road traffic, the decrease of shipping emissions, and the COVID-19 lockdown.

The insights derived from this work contribute to a better understanding of UFP behaviour in urban environments, support evidence-based monitoring recommendations, and inform targeted mitigation measures to improve AQ and protect public health across Europe.

Key words: Ultrafine particles, Particle number size distribution, Positive Matrix Factorisation, Air quality, Urban environment, Traffic emissions

RESUM

VARIABILITAT I CONTRIBUCIÓ DE FONTS DE PARTÍCULES ULTRAFINES EN ENTORNS URBANS EUROPEUS

El material particulat atmosfèric (PM) és un dels principals contaminants de l'aire, amb efectes significatius sobre la salut humana i el clima de la Terra. Aquests efectes depenen, entre altres factors, de la mida de les partícules. Les partícules ultrafines (UFPs, amb diàmetre < 100 nm) representen només una petita fracció de la concentració en massa de PM ($\mu\text{g m}^{-3}$) però constitueixen la major part de la concentració en nombre de partícules (PNC, $\# \text{cm}^{-3}$). Les distribucions de mida de nombre de partícules (PNSDs) es divideixen en tres modes: nucleació (< 25 nm), Aitken (25–100 nm) i acumulació (100–1000 nm).

A causa de la seva mida nanomètrica, les UFPs poden penetrar profundament al sistema respiratori i travessar la barrera alveolar fins al torrent sanguini, arribant a altres òrgans. Tot i això, els estudis de salut encara són limitats per la manca de mètodes de mesura estandarditzats, la poca disponibilitat de dades a llarg termini, l'alta variabilitat espacial i temporal de les UFPs, i les diverses fonts contribuïnt a la PNC en cada regió. El marc regulador per a aquestes partícules encara està en desenvolupament, sense valors límit legalment vinculants. La nova Directiva de Qualitat de l'Aire (Unió Europea) 2024/2881 reconeix per primera vegada les UFP com a contaminants emergents. Tot i que no estableix valors límits, la directiva exigeix la mesura de la PNC i la PNSD a “supersites” urbanes, amb un límit inferior de mida de 10 nm, i a punts calents de contaminació com zones de trànsit intens, aeroports, ports i indústries, així com a àrees rurals.

L'objectiu principal d'aquesta tesi és millorar el coneixement sobre la variabilitat espacial i temporal i les fonts de UFP a ciutat europees. Per això, s'ha compilat, harmonitzat i analitzat dades de PNC i PNSD (2009–2019) de diverses estacions de mesura urbanes a Europa. S'ha avaluat la variabilitat espacial i les tendències temporals a llarg termini, i s'han identificat i quantificat les fonts mitjançant anàlisis de contribució de fonts (SA) amb el model Factorització de Matriu Positiva (PMF). També s'han caracteritzat les emissions d'aviació properes a un gran aeroport europeu i s'ha analitzat l'impacte de la dispersió meteorològica sobre les contribucions de fonts.

Aquest treball ofereix una de les avaluacions més completes fins ara de les concentracions de UFP i de la SA en entorns urbans europeus. L'harmonització de les PNSDs dins del rang de 10–800 nm ha permès comparacions fiables entre estacions i ha minimitzat els artefactes derivats de diferents límits inferiors de detecció. Els resultats mostren un augment progressiu de la PNC del nord cap al sud d'Europa, amb nivells més elevats en estacions de trànsit, confirmant el paper dominant del transport rodat. Les anàlisis de tendència temporal indiquen una disminució clara de les partícules dels models Aitken i acumulació, atribuïda a les polítiques europees de reducció d'emissions del trànsit. En canvi, el mode de nucleació s'ha mantingut estable o lleugerament creixent, influït per processos de nova formació de partícules (NPF) i fonts no vinculades al trànsit com la indústria, transport marítim i aviació.

L'anàlisi de SA mitjançant el model PMF ha identificat deus factors diferenciat que contribueixen a les PNCs, amb el trànsit rodat com a major contribuent, seguint de la nucleació fotoquímica i la calefacció domèstica. Les emissions d'aviació s'han caracteritzat en un estudi a l'Aeroport de Brussel·les, on s'ha observat que les operacions aeronàutiques dominaven els nivells locals de UFPs i que aquestes es mantenen detectables a diversos quilòmetres sotavent. L'aplicació de la normalització per dispersió en un anàlisi de Barcelona (2013–2024) ha millorat els resultats de la SA en separar els canvis impulsats per les emissions dels derivats de la variabilitat meteorològica.

En conjunt, aquesta tesi posa de manifest la necessitat d'una monitorització harmonitzada de les UFPs i d'estratègies de mitigació específiques que abordin tant les fonts primàries com les secundàries. Els resultats proporcionen una base científica sòlida per donar suport al desenvolupament de regulacions fonamentades en l'evidència, orientades a reduir l'exposició de les UFPs i protegir la salut pública a tot Europa.

Paraules clau: Partícules ultrafines, Distribució del nombre de partícules per mida, Factorització de matriu positiva, Qualitat de l'aire, Entorn urbà, Emissions del trànsit

RESUMEN

VARIABILIDAD Y CONTRIBUCIÓN DE FUENTES DE PARTÍCULAS ULTRAFINAS EN ENTORNOS URBANOS EUROPEOS

El material particulado atmosférico (PM) es uno de los principales contaminantes del aire, con efectos significativos sobre la salud humana y el clima de la Tierra. Estos efectos dependen, entre otros factores, del tamaño de las partículas. Las partículas ultrafinas (UFPs, con un diámetro < 100 nm) representan solo una pequeña fracción de la concentración en masa de PM ($\mu\text{g m}^{-3}$), pero constituyen la mayor parte de la concentración en número de partículas (PNC, $\# \text{ cm}^{-3}$). Las distribuciones de tamaño del número de partículas (PNSDs) se dividen en tres modos: nucleación (< 25 nm), Aitken (25–100 nm) y acumulación (100–1000 nm).

Debido a su tamaño nanométrico, las UFPs pueden penetrar profundamente en el sistema respiratorio y atravesar la barrera alveolar hasta el torrente sanguíneo, alcanzando otros órganos. Sin embargo, los estudios sobre salud siguen siendo limitados por la falta de métodos de medición estandarizados, la escasa disponibilidad de datos a largo plazo, la alta variabilidad espacial y temporal de las UFPs, y las diversas fuentes que contribuyen a la PNC en cada región. El marco regulador para estas partículas aún está en desarrollo, sin valores límite legalmente vinculantes. La nueva Directiva de Calidad del Aire (Unión Europea) 2024/2881 reconoce por primera vez las UFPs como contaminantes emergentes. Aunque no establece valores límite, la directiva exige la medición de la PNC y la PNSD en “supersites” urbanos, con un límite inferior de tamaño de 10 nm, en puntos calientes de contaminación como zonas de tráfico intenso, aeropuertos, puertos e instalaciones industriales, así como en áreas rurales.

El objetivo principal de esta tesis es mejorar el conocimiento sobre la variabilidad espacial y temporal y las fuentes de UFPs en las ciudades europeas. Para ello, se han recopilado, armonizado y analizado datos de PNC y PNSD (2009–2019) de varias estaciones de medición urbanas en Europa. Se ha evaluado la variabilidad espacial y las tendencias temporales a largo plazo, y se han identificado y cuantificado las fuentes mediante análisis de contribución de fuentes (SA) con el modelo de Factorización de Matriz Positiva

(PMF). También se han caracterizado las emisiones de aviación cercanas a un gran aeropuerto europeo y se ha analizado el impacto de la dispersión meteorológica sobre las contribuciones de las fuentes.

Este trabajo ofrece una de las evaluaciones más completas hasta la fecha de las concentraciones de UFPs y de la SA en entornos urbanos europeos. La armonización de las PNSDs dentro del rango de 10–800 nm ha permitido comparaciones fiables entre estaciones y ha minimizado los artefactos derivados de diferentes límites inferiores de detección. Los resultados muestran un aumento progresivo de la PNC del norte al sur de Europa, con niveles más elevados en estaciones de tráfico, confirmando el papel dominante del transporte rodado. Los análisis de tendencias temporales indican una clara disminución de las partículas de los modos Aitken y de acumulación, atribuida a las políticas europeas de reducción de emisiones del tráfico. En cambio, el modo de nucleación se ha mantenido estable o ha mostrado un ligero aumento, influido por procesos de nueva formación de partículas (NPF) y por fuentes no relacionadas con el tráfico, como la industria, el transporte marítimo y la aviación.

El análisis de SA mediante el modelo PMF ha identificado diez factores diferenciados que contribuyen a las PNCs, con el tráfico rodado como principal contribuyente, seguido de la nucleación fotoquímica y la calefacción doméstica. Las emisiones de aviación se han caracterizado en un estudio en el Aeropuerto de Bruselas, donde se observó que las operaciones aeronáuticas dominaban los niveles locales de UFPs y que estas se mantenían detectables a varios kilómetros a sotavento. La aplicación de la normalización por dispersión en un análisis de Barcelona (2013–2024) ha mejorado los resultados de la SA al separar los cambios impulsados por las emisiones de los derivados de la variabilidad meteorológica.

En conjunto, esta tesis pone de manifiesto la necesidad de una monitorización armonizada de las UFPs y de estrategias de mitigación específicas que aborden tanto las fuentes primarias como las secundarias. Los resultados proporcionan una base científica sólida para apoyar el desarrollo de regulaciones fundamentadas en la evidencia, orientadas a reducir la exposición a las UFPs y proteger la salud pública en toda Europa.

Palabras clave: Partículas ultrafinas, Distribución del número de partículas por tamaño, Factorización de matriz positiva, Calidad del aire, Entorno urbano, Emisiones del tráfico

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ABBREVIATIONS AND CHEMICAL SPECIES

Abbreviations

AE	Aethalometer
AQ	Air Quality
a.s.l.	Above Sea Level
BC	Black Carbon
BLH	Boundary Layer Height
C3S	Copernicus Climate Change Service
CCN	Cloud Condensation Nuclei
CDNC	Cloud Droplet Number Concentration
CI-API-ToF	Chemical Ionisation Atmospheric Pressure interface Time of Flight Mass Spectrometer
CPC	Condensation Particle Counter
CS	Condensation Sink
CTM	Chemical Transport Model
DC	Dispersion Corrected
DISP	Displacement
DMA	Differential Mobility Analyser
DMPS	Differential Mobility Particle Sizer
DPF	Diesel Particulate Filter
E	East
EC	European Commission
ECMWF	European Centre for Medium-range Weather Forecasts
EEA	European Environment Agency
ERF	Effective Radiative Forcing
EU	European Union
GHG	Greenhouse Gases
GRIMM	Grimm Aerosol Technik
LTO	Landing and Takeoff Operations
MAAP	Multi-Angle Absorption Photometer
ME-2	Multilinear Engine 2
MPSS	Mobility Particle Size Spectrometer

N	North
NPF	New Particle Formation
OPC	Optical Particle Counter
P	Pressure
PCA	Principal Component Analysis
PM	Particulate Matter
PM₁	Particulate Matter of aerodynamic diameter below 1 µm
PM_{2.5}	Particulate Matter of aerodynamic diameter below 2.5 µm
PM₁₀	Particulate Matter of aerodynamic diameter below 10 µm
PMF	Positive Matrix Factorisation
PN	Particle Number
PNC	Particle Number Concentration
PNSD	Particle Number Size Distribution
PSM	Particle Size Magnifiers
R	Rain
RAD	Solar Radiation
RB	Regional Background
S	South
ss	Statistical Significance
SA	Source Apportionment
SIA	Secondary Inorganic Aerosol
SMPS	Scanning Mobility Particle Sizer
SU	Stockholm University
SUB	Suburban Background
SVOC	Semi-Volatile Organic Compound
T	Temperature
TDMA	Tandem Differential Mobility Analyser
TDMPS	Twin Differential Mobility Particle Sizer
TPN	Total Particle Number
TR	Traffic
TROPOS	Leibniz Institute for Tropospheric Research
TSI	TSI Incorporated
TSMPS	Twin Scanning Mobility Particle Sizer
UB	Urban Background
UC	Uncorrected

UFP	Ultrafine Particle
UHEL	University of Helsinki
UK	United Kingdom
US	United States
US EPA	US Environmental Protection Agency
UV	Ultraviolet
VC	Ventilation Coefficient
VMM	Flanders Environment Agency
VOC	Volatile Organic Compound
W	West
WD	Wind Direction
WS	Wind Speed
WHO	World Health Organization

Chemical species

As	Arsenic
BaP	Benzo[a]pyrene
Cd	Cadmium
CH₄	Methane
C₆H₆	Benzene
CO	Carbon monoxide
CO₂	Carbon dioxide
H₂O	Water vapour
NH₃	Ammonia
Ni	Nickel
NO₂	Nitrogen dioxide
NO_x	Nitrogen oxides
N₂O	Nitrous oxide
O₃	Ozone
Pb	Lead
SO₂	Sulphur dioxide

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1 INTRODUCTION

1.1 Air quality and pollutants

The Earth's atmosphere is a complex mixture of gases and particles in all states of matter, influenced by both natural processes and human activities. In the last decades, the rapid growth of population, industrialisation, urban expansion, and the associated increase in energy production based on fossil fuel combustion, among other factors, have altered the composition of the atmosphere. Pollutants can be emitted into the atmosphere either by anthropogenic sources, such as transport, industry, homes, agriculture, and waste management, or by natural sources, such as sea salt, wildfires, mineral dust, and volcanic emissions, among others. Once released into the atmosphere, pollutants can remain for varying durations and interact chemically and physically, leading to the formation of secondary pollutants.

All the above anthropogenic and natural sources contribute to atmospheric contamination at two general levels: climate change and air pollution.

In the first case, referring to climate change, greenhouse gases (GHGs) such as carbon dioxide (CO₂), methane (CH₄), water vapour (H₂O), and tropospheric ozone (O₃) are responsible for most of the global warming currently affecting the atmosphere (IPCC, 2021). Furthermore, aerosols may also have a climate impact in two main ways: i) by directly scattering or absorbing radiation, resulting in cooling and warming effects, respectively; and ii) by influencing cloud condensation nuclei (CCN), thereby affecting CCN density (and thus their light-scattering potential), cloud lifetime, and precipitation patterns (IPCC, 2021).

On the other hand, there are other pollutants, also emitted by the above sources, that cause air quality (AQ) impairment. These AQ pollutants can be broadly classified based on their physical state and chemical composition. Gaseous pollutants include sulphur dioxide (SO₂), nitrogen dioxide and oxides (NO₂ and NO_x), carbon monoxide (CO), benzene (C₆H₆), and O₃.

Particulate pollutants, commonly referred to as aerosols, are solid, liquid, or mixed-phase particles suspended in the air (Seinfeld and Pandis, 2016). Within this category, suspended particulate matter (PM) refers specifically to the particles themselves and are categorised by size. Particle sizes can range from a few nanometres to several hundred micrometres. They are typically categorised as ultrafine, fine, and coarse particles, corresponding to aerodynamic diameters below 100 nm, 2.5 μm , and 10 μm , respectively. Aerosol mass concentrations are usually expressed in $\mu\text{g m}^{-3}$, commonly in terms of PM_{10} , $\text{PM}_{2.5}$, and PM_1 , which refer to particles with aerodynamic diameters smaller than 10, 2.5, and 1 μm , respectively.

In addition to gaseous and particulate pollutants, trace components of PM are monitored as indicators of exposure to harmful substances, including C_6H_6 , lead (Pb), arsenic (As), cadmium (Cd), nickel (Ni), and benzo[a]pyrene (BaP). Although they represent only a small fraction of the total PM mass, their presence is critical due to their high toxicity even at low concentrations.

The monitoring of these pollutants is a significant public health concern. According to the World Health Organization (WHO), air pollution is the second leading risk factor for noncommunicable diseases, with cardiovascular conditions, strokes, pulmonary diseases, and lung cancer being the most common causes of premature death attributable to it (WHO, 2024). In 2019, ambient air pollution in both urban and rural areas was estimated to cause 4.2 million premature deaths worldwide, primarily due to exposure to fine PM, which contributes to cardiovascular and respiratory diseases, as well as cancers. Of these premature deaths related to outdoor air pollution, 68% were due to ischaemic heart disease and stroke, 14% to chronic obstructive pulmonary disease, 14% to acute lower respiratory infections, and 4% to lung cancers.

The European Environment Agency (EEA) estimated that exposure to $\text{PM}_{2.5}$, O_3 , and NO_2 was responsible for 253,000, 52,000, and 22,000 premature deaths, respectively, in the European Union (EU) in 2022 (EEA, 2024). However, between 2005 and 2022, the number of deaths in the EU attributable to $\text{PM}_{2.5}$ decreased by 45%, bringing the EU closer to achieving the 55% reduction target outlined in the European Green Deal's zero

pollution action plan for 2030, as set by the European Commission (EC) in its policy framework (EC, 2021).

In addition to the climate and AQ effects, air pollution may also affect ecosystems, the durability of materials, and visibility (Watson, 2002; Fowler et al., 2009; Rao et al., 2014).

The above AQ pollutants are referred to as “criteria pollutants” because evidence provided by the WHO is sufficient to support the establishment of limit or target values, which are included in AQ standards and must be not exceeded by Member States of the EU. However, there are other additional pollutants that, while not formally regulated as criteria pollutants, are crucial for a scientifically advanced AQ assessment aimed at mitigating pollution and protecting public health. These include pollutants such as ultrafine particles (UFP), black carbon (BC), volatile organic compounds (VOCs), ammonia (NH₃), and PM speciation, among others (WHO, 2021a). As it will be shown in subsequent sections, many of these non-criteria pollutants are included in the new AQ standards in Europe, without specific limit or targeted values.

1.2 Ultrafine particles and particle number size distribution

Among the various pollutants that compromise AQ, UFPs have received growing scientific and policy interest due to their unique physicochemical characteristics and potential health impacts. UFPs are defined as particles with diameters less than or equal to 100 nm. While they contribute only a small fraction to PM mass concentrations (measured in terms of mass per unit volume, $\mu\text{g m}^{-3}$), they account for a substantial proportion of particle number concentration (PNC, measured in particles per cubic centimetre, $\# \text{ cm}^{-3}$). The term “nanoparticle” refers to the same size range but is typically used in materials science, whereas “UFP” is the preferred term in atmospheric sciences. UFPs are recommended to be measured for a size range with a lower limit of 10 nm and no restrictions on the upper size limit (WHO, 2021a). Since the number fraction of particles greater than 100 nm generally comprises 10–30% compared to that <100 nm (typically around 80%, Baldauf et al., 2016; Sun et al., 2019), the use of PNC, defined as the number of particles related to the unit volume of the carrier gas (CEN, 2024), without

an upper size limit is considered a reasonable approximation for the number concentration of UFP (<100 nm). Consequently, PNC is used as a surrogate for UFPs, and in this thesis, the two terms will be used interchangeably.

One of the most relevant parameters in this context is the particle number size distribution (PNSD), defined by the frequency distribution of PNC as a function of the logarithm of particle size, such that the area under the distribution curve between two sizes corresponds to the number concentration within that size range (CEN, 2020). In practice, PNSD is determined by measuring PNC across discrete size intervals.

1.2.1 Particle size ranges

Particles in the atmosphere are influenced by emission, formation, and transformation processes, and they exhibit a size distribution comprising the following general size modes (Figure 1.1):

- Nucleation mode (<25 nm): This mode is dominated by new particle formation (NPF) processes (Seinfeld and Pandis, 2016), which occur through photochemical reactions enhanced by high solar radiation (RAD) and wind speed (WS), low relative humidity (RH), a low condensation sink (CS, due to a limited surface area on pre-existing particles), and the presence of precursor gases such as SO₂ and NH₃ (Kulmala and Kerminen, 2008), as well as VOCs that favour the growth of newly generated particles (Ehn et al., 2014). Furthermore, in urban environments, road traffic exhaust emissions are major contributors to nucleation mode particles, which are either emitted directly from engines or formed through nucleation in the atmosphere immediately after the release of semi-volatile organic compounds (SVOCs) from tailpipes (Harrison et al., 2011; Rönkkö and Timonen, 2019).
- Aitken mode (25–100 nm): This mode includes particles that are either directly emitted from combustion processes or formed through the growth of pre-existing particles via mechanisms such as gas-to-particle conversion, coagulation, and condensation of nucleation mode particles (Seinfeld and Pandis, 2016). In urban areas, Aitken mode particles are predominantly associated with traffic emissions from both gasoline and diesel vehicle (Charron and Harrison, 2003).

- Accumulation mode (100–1000 nm): This mode is dominated by Aitken mode particles that have grown through coagulation of pre-existing particles and condensation of SVOCs, as well as SO₂- and NO₂-derived acids (Seinfeld and Pandis, 2016). It includes both primary particles from combustion processes, such as soot, and secondary particles derived from precursor gases. These particles are found in lower PNCs in the atmosphere but higher mass concentrations.
- Coarse mode (>2.5 µm): This mode is mainly composed of primary particle generated by mechanical processes, including soil dust, marine aerosols, biogenic debris, tyre and brake wear particles, and road dust. Additionally, secondary coarse particles may arise from interactions between gaseous precursors with mineral matter or sea salt. Typically, coarse mode particles account for approximately 95% of the total mass concentration, but only around 5% of the PNC (Seinfeld and Pandis, 2016).

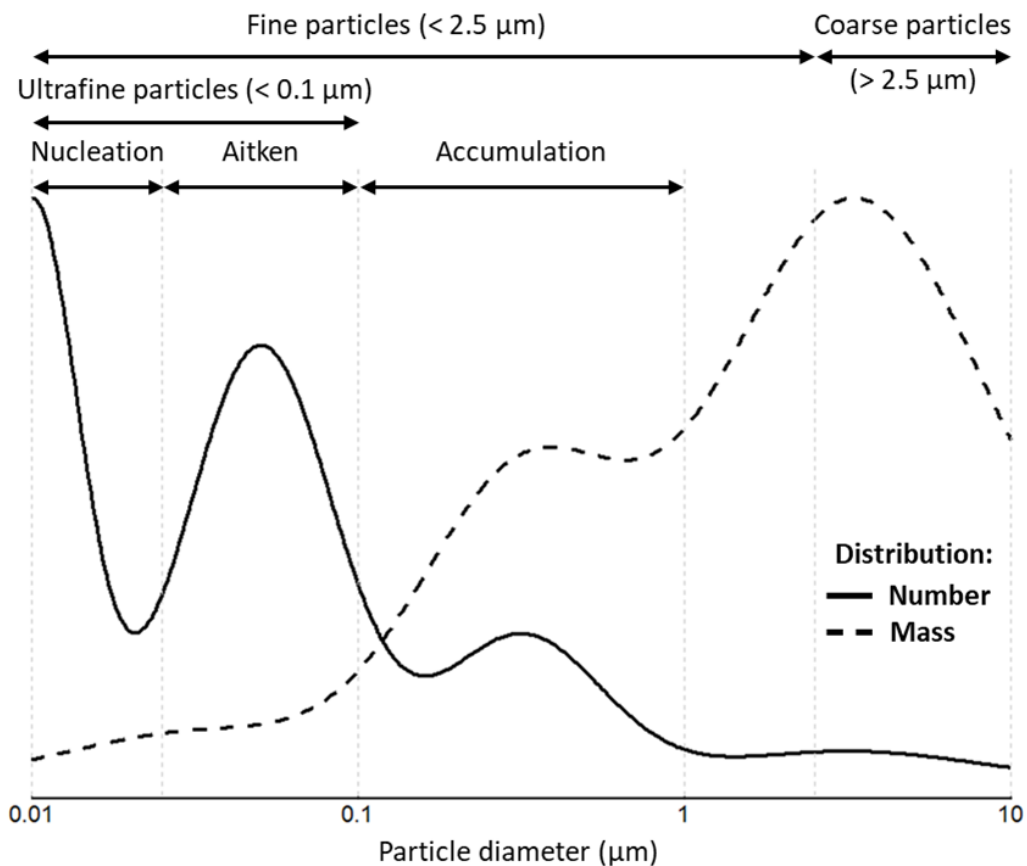


Figure 1.1: Simplified schematic of the particle mass (dashed line) and number (solid line) distribution for a typical urban aerosol, including particle size classification names.

1.2.2 Origin of ultrafine particles

Atmospheric UFPs can also be classified according to their formation processes into primary or secondary particles. Primary particles are directly emitted as particles into the atmosphere from the sources, while secondary particles are formed in the atmosphere by gas-to-particle conversion processes (Seinfeld and Pandis, 2016). Global PNCs are dominated by anthropogenic secondary aerosols (Gordon et al., 2017), whereas global annual mass fluxes of PM are dominated by natural primary particles (Gieré and Querol, 2010).

In urban environments, primary UFPs are predominantly anthropogenic and produced by combustion processes in the nucleation and Aitken modes, mainly from vehicle tailpipe emissions (Hama et al., 2017). Industrial facilities involving high temperatures processes, such as power generation plants and other combustion related industries, as well as biomass burning, also emit primary particles (Kumar et al., 2014).

Secondary particles are mainly formed from gaseous precursors (e.g., NO_x, SO₂, NH₃, amines, VOCs, among others), which undergo atmospheric chemical reactions that results in particle formation (Kulmala et al., 2004).

The main mechanisms through which secondary UFPs are formed is NPF (Figure 1.2), which involves the formation of clusters composed of precursor vapor molecules through photochemical reactions (Kulmala et al., 2004). These molecular clusters have very short lifetimes, as they are constantly colliding to larger particles that absorb them (scavenging). Under favourable conditions, clusters can be stabilised by agents (amines, NH₃, and VOCs), allowing them to reach a critical size and nucleate to form particles (Kirkby et al., 2011; Kulmala et al., 2013). Once formed, these particles must be activated to enhance their growth, typically via condensation of VOCs (Kulmala et al., 2013). NPF events are enhanced by several atmospheric conditions (Kulmala et al., 2004; Kulmala and Kerminen, 2008; Hamed et al., 2011; Kerminen et al., 2018): i) high RAD, which promotes the production of oxidants (OH and O₃) that convert gaseous precursors into low-volatility compounds; ii) high WS, which can transport newly formed particles away from their source regions, potentially influencing the spatial distribution of NPF events; iii) low RH, which reduces scavenging of precursors by water or existing particles; iv)

low CS, which minimises the loss of molecular clusters to pre-existing particles; and v) the presence of precursor gases, such as SO₂ and NH₃.

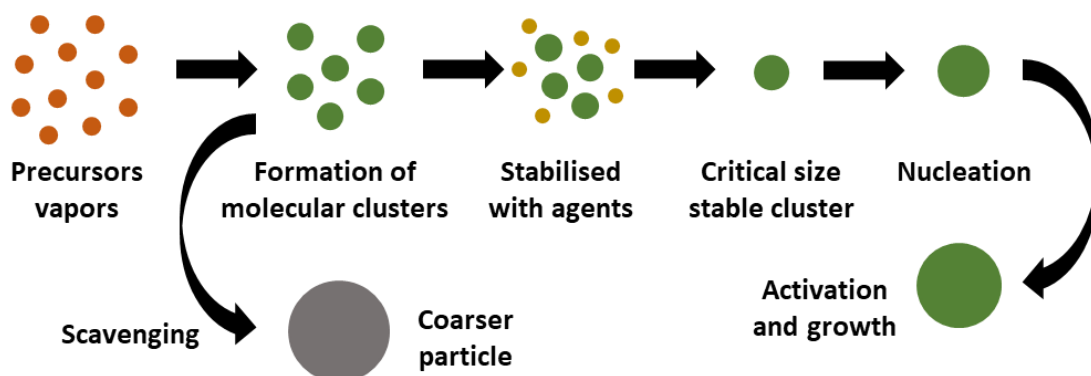


Figure 1.2: Schematic description of the NPF mechanisms.

These NPF events are commonly observed at the regional scale, and it is accepted that 50–60% of the global CCN arise from NPF and subsequent growth (Gordon et al., 2017). However, such events may also occur in urban environments, where NPF is influenced by several sources and processes (Dall’Osto et al., 2011). These include emissions from road traffic, in which particles are nucleated after the emission of SVOCs present in diesel exhaust into the atmosphere, as well as other precursors from shipping activities and power plants that emit plumes rich in precursor gases, and photochemically induced nucleation at regional-scale, highly influenced by O₃ (Harrison et al., 2011; Junkermann et al., 2011; Ehn et al., 2014; Kalkavouras et al., 2020; Damayanti et al., 2023).

While secondary formation processes are significant in urban environments, primary UFPs are very relevant in terms of concentrations. According to the RI-URBANS (2025) European Emission Inventory for 2019, primary UFP emissions are dominated by shipping emissions ($950 \cdot 10^{24}$ particles yr⁻¹), followed by road transport, energy and industry, and aviation ($580 \cdot 10^{24}$, $470 \cdot 10^{24}$, and $250 \cdot 10^{24}$ particles yr⁻¹, respectively) (Figure 1.3). This ranking of emission sectors differs markedly from that of PM_{2.5}.

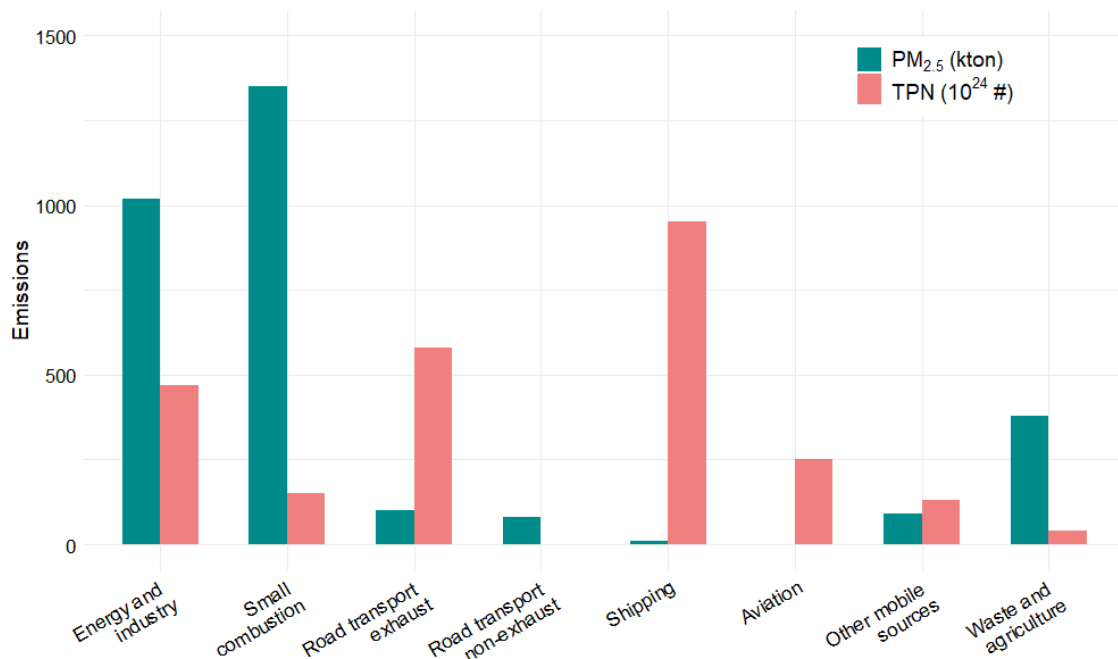


Figure 1.3: Comparison between PM_{2.5} (mass in kilotons) and total particle number (TPN, in 10²⁴ number of particles) emissions across different sectors in 2019 (total for Europe). Adapted from RI-URBANS (2025).

When considering the source contributions to urban ambient PNC, the ranking of contributing sources differs markedly from that of emission inventories for primary UFP. This discrepancy is primarily due to: i) the proximity of road traffic to monitoring sites and citizens, and ii) the relevance of secondary NPF. Thus, source apportionment (SA) studies are usually carried out to determine source/processes contributions to UFP.

1.2.3 Source apportionment of ultrafine particles

Hopke et al. (2022) reviewed the SA literature of ambient UFP based on PNSD measurements and identified 55 peer-review articles reporting SA of PNSD in 102 locations or time periods. While almost all of these studies were performed using Positive Matrix Factorisation (PMF, Paatero and Tapper, 1994), other methods have also been applied in SA, including Principal component analysis (PCA) and k-means clustering.

PCA is a multivariate statistical technique that reduces the dimensionality of PNSD data by grouping size bins that co-vary over time into components. These components can

then be interpreted as potential sources, but PCA does not provide quantitative source contributions. It was first applied in this context by Thurston and Spengler (1985) and has since been used in several studies (Wehner and Wiedensohler, 2003; Oliveira et al., 2009; Pey et al., 2009; Cusack et al., 2013; Khan et al., 2015; among others).

K-means clustering partitions PNSD data into cluster characterised by specific emission or formation process, enabling the identification of temporal and spatial trends in the size distribution. This method has also been applied in various studies (Dall'Osto et al., 2011, 2019; Wegner et al., 2012; Brines et al., 2014; Brines et al., 2015; Chen et al., 2021; among others).

Additionally, Rodríguez and Cuevas (2007) developed a methodology based on correlations with BC to separate PNCs affected by fresh vehicle exhaust emissions into two components: one accounting for primary particles directly emitted in the particle phase or nucleating immediately after the emissions, and the other representing secondary UFPs from NPF or other atmospheric processes.

PMF is the most widely used receptor modelling method for identifying and quantifying the sources of PNSD. It is a multivariate factor analysis technique that assumes the observed aerosol matrix X (with n observations and m size bins) can be represented as the product of a factor profile matrix (F) and a factor contribution matrix (G), for a user-defined number of factors (interpreted as sources). This is mathematically expressed as:

$$x_{ij} = \sum_{\{k=1\}}^p g_{ik} f_{kj} + e_{ij} \quad (1)$$

where p is the total number of factors, x_{ij} is the measured particle number concentration in the j th size bin of the i th sample, f_{jk} is the concentration profile of the j th size bin for the k th source, g_{ik} is the contribution of the k th source to the i th sample, and e_{ij} represents the residual or error term for the i th sample and j th size bin (i.e., the adjustment error).

Based on the SA results reviewed by Hopke et al. (2022), the most commonly identified source categories in urban environments have been generally characterised as:

- Nucleation: Characterised by a single mode in the PNSD with a peak typically between 10–25 nm. It often presents peaks during the morning and evening rush hours due to the emission of primary SVOCs from vehicles, which nucleate as they dilute and cool (Harrison et al., 2011). A midday peak is also common, driven by NPF under high photochemical activity.
- Road traffic: Two distinct traffic-related factors are frequently reported. One shows a peak around 30–35 nm, typically linked to spark-ignition vehicle emissions (Johnson et al., 2005; Kittelson, 1998), while the other peaks around 60–80 nm and is more characteristic of diesel engine exhaust (Dua et al., 1999; Kittelson, 1998).
- Heating: This factor typically exhibits a mode around 90–100 nm and is linked to combustion from various fuels used for building heating (Chang et al., 2004; Chandrasekaran et al., 2011).
- O₃-Rich: Displays variable PNSD patterns with multiple modes. The co-variation with O₃ and midday peaks suggest that this factor may reflect particles grown through the condensation of secondary material, including both organic and inorganic compounds (Ogulei et al., 2007; Kasumba et al., 2009; Liu et al., 2014; Squizzato et al., 2019).
- Secondary Inorganic Aerosol (SIA): Characterised by particle modes typically above 100 nm, with winter contributions associated with secondary nitrate and summer peaks linked to secondary sulphate (Kasumba et al., 2009; Squizzato et al., 2019).
- Other sources: Generally dominated by particles with modes above 100nm. This category includes a range of sources such as biomass burning, urban background, industrial emissions, mixed sources, dust, and unidentified contributors.

Overall, most studies reviewed by Hopke et al. (2022) reported that road traffic is the dominant contributor to PNCs in urban areas.

1.2.4 Dispersion of ultrafine particles

The concentration and spatial distribution of UFPs in urban environments are influenced not only by the atmospheric emissions of primary UFP and gaseous precursors but also by meteorological conditions, geographic features, and urban structural characteristics, which can promote NPF, and either the accumulation or dispersion of particles (Kumar et al., 2014).

Proximity to major sources is one of the primary determinants of local UFP concentrations. Areas with high traffic density, industrial activities, or those located near airports or harbours tend to exhibit elevated PNCs (de Jesus et al., 2019). However, the layout of buildings, streets, and open spaces also affects airflow patterns and can significantly influence UFP dispersion dynamics. In urban street canyons (narrow streets bordered by high-rise buildings), the combination of high UFP emissions and limited dilution often leads to the trapping of pollutants near ground level, making these locations hotspots for UFP accumulation (Weber et al., 2013). In contrast, open areas such as parks or airports promote greater air circulation and contribute to the dispersion of particles (Zhu et al., 2021).

Geographical features further contribute to the variability of UFP concentrations. Cities situated in valleys often experience stable atmospheric conditions and reduced vertical mixing, which can lead to the accumulation of pollutants near the surface. Temperature inversions, where a layer of cold air is trapped beneath a layer of warmer air, also inhibit or strongly reduce vertical mixing, effectively confining pollutants, including UFPs, within the inversion layer close to the ground (EEA, 2020). Moreover, local terrain can modify wind circulation patterns, either accumulating particles in certain areas or aiding their transport and dispersion.

Variability in PNCs is also strongly governed by meteorological processes, primarily influenced by (Andronache, 2004; Kumar et al., 2011; Wang et al., 2023): i) solar radiation, which increases the planetary boundary layer height (BLH), enhancing particle dispersion, and also drives photochemical reactions that can lead to NPF; ii) precipitation, which contributes to the scavenging and removal of particles; and iii) WS, which promotes dispersion under strong winds and allows accumulation under calm conditions.

1.2.5 Health impact of ultrafine particles

Cassee et al. (2019) and WHO (2021a) reported a body of evidence indicating adverse health effects derived from the exposure to high ambient concentrations of UFPs, particularly among individuals living in densely populated urban areas. However, they also noted that results are inconsistent and highlight the need for further epidemiological studies.

Epidemiological studies have evidenced that exposure to aerosols is associated with an increased risk of respiratory and cardiovascular diseases, as well as premature mortality (WHO, 2024). Due to their small size, UFPs have been suggested to be more toxic than larger particles (Davidson et al., 2005).

Peters et al. (1997) conducted the first epidemiological study demonstrating short-term effects of ambient UFPs, showing that PNCs were associated with decreases in lung function, consistent with a cumulative response over several days. Since then, both toxicological and epidemiological studies have further investigated the health impacts of UFPs.

Epidemiological evidence has linked PNC exposure to increased cardiovascular mortality (Atkinson et al., 2010) and has highlighted the high potential of UFPs for deep lung deposition (Oberdörster et al., 2005). Due to their nano size, UFPs can not only penetrate the deepest parts of the respiratory tract but also to translocate through the alveolar barrier into the bloodstream, reaching other organs throughout the body from the circulatory system (Figure 1.4; Oberdörster et al., 2005; Peters et al., 2006; Kreyling et al., 2014; Cassee et al., 2019). Other studies have provided evidence of UFP translocation into the brain via the olfactory nerve in the nose, and have shown that UFPs can cross cell membranes through mechanisms other than phagocytosis (Oberdörster et al., 2004; Genc et al., 2012; among others).

Multiple studies have also reported a positive association between UFP exposure and increased premature mortality (e.g., Wichmann et al., 2000; Ibalá-Mulli et al., 2004; Tobías et al., 2018; Schwarz et al., 2023), further underscoring the serious health risks posed by UFPs.

Toxicological studies have shown that UFPs can induce oxidative stress, inflammation and cellular damage. These effects are strongly influenced by the chemical composition, size, and source of the particles, as well as by atmospheric conditions (Li et al., 2003; Jalava et al., 2007; Steenhof et al., 2011).

However, despite the growing evidence of the health effects of UFPs, long-term exposure assessments remain challenging due to the lack of standardised monitoring methods, limited long-term data, the high spatial and temporal variability of UFPs, and the diverse sources contributing to PNCs in each region, highlighting the need for improved regulations and further research (Cassee et al., 2019; Rivas et al., 2021).

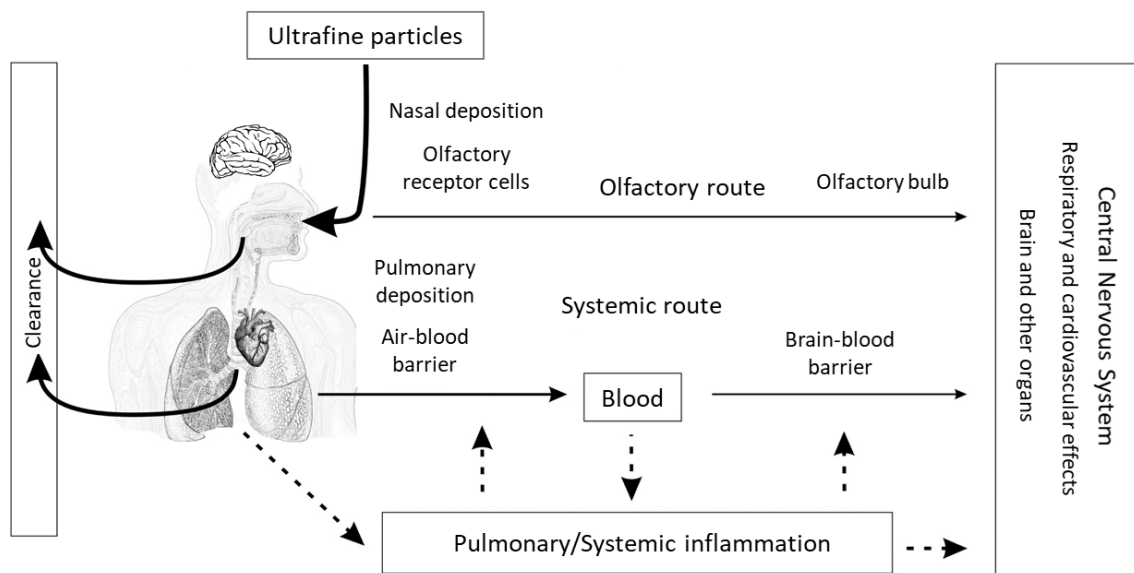


Figure 1.4: Main translocation routes and pathways of UFPs in the human body that may lead to the central nervous system. Adapted from Heusinkveld et al. (2016).

1.2.6 Impact of ultrafine particles on climate and clouds

Beyond their implications for human health, UFPs contribute significantly to atmospheric processes, particularly cloud formation and radiative forcing, playing a key role in climate dynamics.

A major climatic effect of aerosols is their ability to scatter and absorb solar radiation, thereby contributing to radiative forcing, which can result in either a warming or cooling effect depending on their composition (Seinfeld & Pandis, 2016; Qin-Qin Li et al., 2023). Particles containing BC, which strongly absorbs solar radiation, can lead to localised direct atmospheric warming, particularly in densely populated areas or regions affected by forest fires or biomass combustion (Bond et al., 2013). However, in general, aerosols exert a net direct negative radiative forcing by reflecting incoming solar radiation, thereby contributing to a cooling effect on the climate system (IPCC, 2021). In addition to this direct cooling effect, aerosols also contribute an indirect cooling effect by enhancing the scattering of light by cloud droplets. These droplets form around CCN, which are hygroscopic aerosols capable of growing in size through the condensation of water vapor.

According to IPCC (2021), GHGs contributed a strong positive effective radiative forcing (ERF) estimated at $+3.84 \text{ W m}^{-2}$, whereas aerosols exert a substantial net negative forcing of -1.06 W m^{-2} (Figure 1.5). The total anthropogenic radiative forcing was estimated at $+2.72 \text{ W m}^{-2}$ in 2019 relative to 1750, leading to a net warming global effect on the climate system. The cooling effect of aerosols reached -0.22 W m^{-2} from aerosol-radiation interactions and -0.84 W m^{-2} from aerosol-cloud interactions. Among aerosol types, BC contributed a warming effect ($+0.30 \text{ W m}^{-2}$), while sulphate (-0.44 W m^{-2}), organic carbon (-0.09 W m^{-2}), and nitrate (-0.11 W m^{-2}) contributed to cooling (IPCC, 2021).

UFPs can influence the radiative forcing both directly and indirectly. In the first case, by producing particles able to grow and scatter radiation, or in some cases by absorbing it. The indirect effect is caused by acting as CCN, thereby affecting cloud microphysical properties, formation, and lifetime, and contributing to radiative forcing (Figure 1.6). Nucleated particles during NPF events must undergo atmospheric processes after their activation to grow in size, primarily through the condensation of low-VOCs onto their surface or by coagulation with other particles (Ehn et al., 2014). Particles that reach a critical size and possess specific chemical composition and hygroscopic properties can act as CCN (Petters and Kreidenweis, 2008), typically within the size range of 60–115 nm (Hammer et al., 2014). This process alters cloud properties by increasing cloud droplet number concentration (CDNC) and density, and reducing individual droplet size,

thereby increasing the total cloud droplet surface area and cloud optical thickness, which enhances cloud reflectivity, or albedo (Twomey, 1977).

CCN can also modify precipitation. The reduction in size of the droplets slows their coalescence, potentially suppressing drizzle and rainfall, particularly in shallow or warm clouds (Andreae & Rosenfeld, 2008). Reduced precipitation increases cloud cover and lifetime, further enhancing cloud albedo (Figure 1.6). This higher albedo, due to both increased cloudiness and longer-lived clouds, can intensify the cooling effect associated with negative radiative forcing (Rosenfeld et al., 2014). Moreover, suppressed early precipitation can lead to greater water accumulation within clouds, potentially triggering extreme weather events such as intense rainfall or hailstorms (Su et al., 2020; Shu et al., 2022).

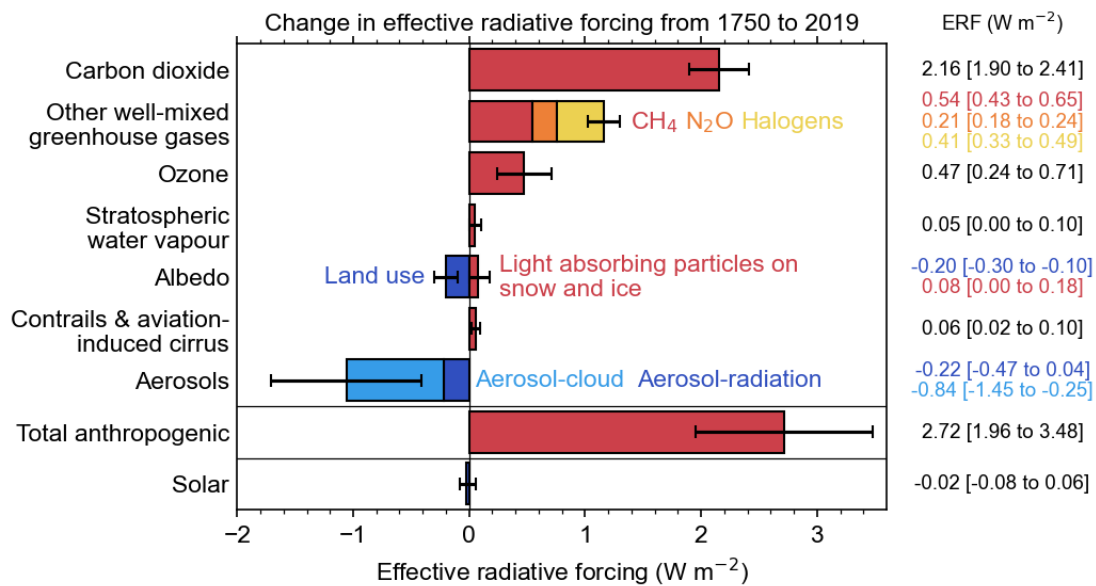


Figure 1.5: Change in ERF from 1750 to 2019 by contributing forcing agents. Other well-mixed greenhouse gases are broken down into contributions from CH₄, nitrous oxide (N₂O) and halogenated compounds. Surface albedo is broken down into land-use changes and light-absorbing particles on snow and ice. Aerosols are broken down into contributions from aerosol-cloud and aerosol-radiation interactions. Solid bars represent best estimates, and very likely (5–95%) ranges are given by error bars. Source: IPCC (2021).

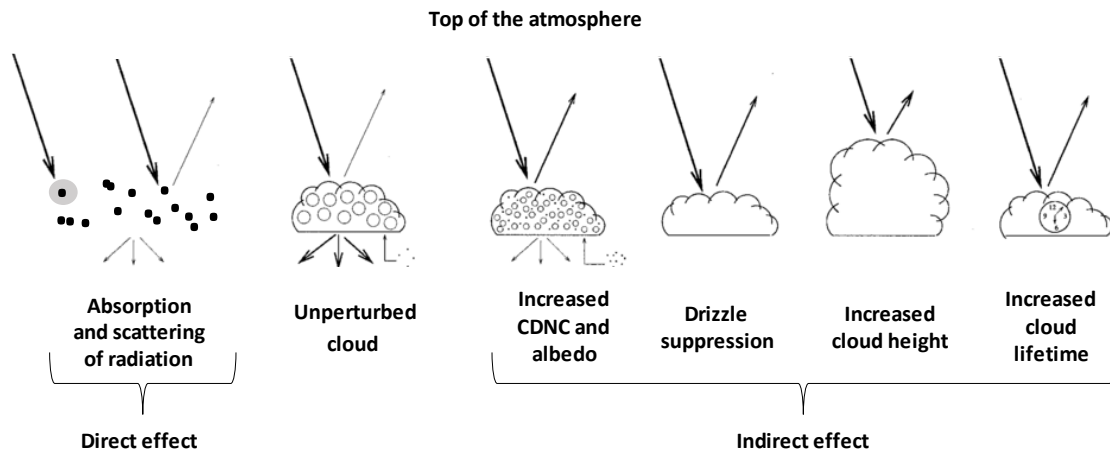


Figure 1.6: Schematic representation of the aerosol effects on clouds. Adapted from Haywood and Boucher (2000).

In addition to the health, climate and cloud effects, one of the most immediate and observable impacts of aerosols is the reduction of atmospheric visibility due to the scattering and absorption of solar radiation, which, in highly polluted environments, can reduce the visibility to just a few meters (Wu et al., 2005; Deng et al., 2008). Additionally, particles may deposit on buildings and ecosystems, leading to reduced durability of exposed construction materials, as well as eutrophication and degradation of ecosystems (Torfs and Van Grieken, 1997; Janhäll, 2015).

1.3 Air quality standards and guidelines

As previously stated, air pollution remains a major environmental and public health risk. To abate the impacts derived from this, regulation and mitigation strategies are crucial, aiming to reduce emissions, improve AQ, and protect human health.

The WHO and the EC are two key bodies responsible for developing guidelines and standards targeting air pollutants. The WHO provides AQ guidelines based on scientific evidence on health effects, offering concentration thresholds for key pollutants and serving as a reference for national and international regulatory frameworks (WHO,

2021a). While WHO guidelines are non-binding, the EC sets legally binding EU AQ standards for member states through directives.

In 2004 and 2008, the Ambient Air Quality Directives 2004/107/EC and 2008/50/EC (EC, 2004 and 2008) consolidated existing AQ legislation and established limit and target values for a range of air pollutants, as well as requirements for AQ assessment and action plans. The publication of updated AQ guidelines by the WHO in 2021, which included more restrictive values based on the latest scientific evidence (WHO, 2021a; WHO, 2021b), prompted a revision of the EU directives. In 2024, the EC published the new Ambient Air Quality Directive (EU) 2024/2881 (EC, 2024), marking an important step toward strengthening AQ monitoring and enforcement, promoting more ambitious national and local measures, and achieving cleaner air across Europe.

1.3.1 Overview of standards for regulated pollutants

The EU directives define AQ standards for a set of regulated pollutants known to pose significant risks to human health and the environment. These include PM₁₀, PM_{2.5}, NO₂, SO₂, CO, O₃, C₆H₆, Pb, As, Cd, Ni, and BaP. Each pollutant is assigned either a limit value, which is legally binding and must not be exceeded, or a target value, which is also legally defined but allows greater flexibility. Concentrations are reported based on different averaging periods, including annual mean, daily (24-hour) mean, hourly (1-hour) mean, or maximum daily 8-hour mean, depending on the pollutant.

In the Ambient Air Quality Directive (EU) 2024/2881, standards were updated to better reflect current scientific evidence, lowering existing limit values and introducing additional ones, particularly for PM and gaseous pollutants, to more closely align with the WHO AQ guidelines. Table 1.1 provides a comparison of the AQ standards set in the 2008 and 2024 EU directives alongside the WHO 2021 guideline values.

Additionally, the new 2024 EU directive enhanced public access to AQ information by requiring real-time user-friendly data presentation. It also established a network of supersites for advanced AQ monitoring, designed to measure both regulated and emerging pollutants and to provide high-resolution data. Emerging pollutants were newly integrated in the directive for obligatory monitoring, though no limit or target values have been set, including BC, elemental carbon, NH₃, the oxidative potential of PM, and UFPs.

Their inclusion reflects growing recognition of their health relevance and the need for improved data collection and scientific understanding.

Table 1.1: Standards for regulated pollutants established in the EU Ambient Air Quality Directives (EC, 2008; EC, 2024) and the WHO air quality guidelines (WHO, 2021a). WHO values represent non-binding recommended thresholds (in black), while EU values are either legally binding limit values (in red) or legally defined target values (in black).

Pollutant	Average period	EU Directive (2008)	WHO (2021)	EU Directive (2024)
PM _{2.5} , µg m ⁻³	24-hour	–	25 (3 days yr ⁻¹)	25 (18 days yr ⁻¹)
	Annual	25	5	10
PM ₁₀ , µg m ⁻³	24-hour	50 (35 days yr ⁻¹)	45 (3 days yr ⁻¹)	45 (18 days yr ⁻¹)
	Annual	40	15	10
NO ₂ , µg m ⁻³	1-hour	200 (18 h yr ⁻¹)	–	200 (3 h yr ⁻¹)
	24-hour	–	25 (3 days yr ⁻¹)	50 (18 days yr ⁻¹)
	Annual	40	10	20
SO ₂ , µg m ⁻³	1-hour	350 (3 h yr ⁻¹)	–	350 (3 h yr ⁻¹)
	24-hour	125 (24 days yr ⁻¹)	40 (3 days yr ⁻¹)	50 (18 days yr ⁻¹)
	Annual	–	–	20
CO, mg m ⁻³	Max. daily 8-hour mean	10	–	10
	24-hour	–	4 (3 days yr ⁻¹)	4 (18 days yr ⁻¹)
O ₃ , µg m ⁻³	Max. daily 8-hour mean	120 (25 days yr ⁻¹)	100 (3 days yr ⁻¹)	120 (18 days yr ⁻¹)
C ₆ H ₆ , µg m ⁻³	Annual	5	–	3.4
Pb, µg m ⁻³	Annual	0.5	–	0.5
As, ng m ⁻³	Annual	6	–	6
Cd, ng m ⁻³	Annual	5	–	5
Ni, ng m ⁻³	Annual	20	–	20
BaP, ng m ⁻³	Annual	1	–	1

1.3.2 Guidelines for ultrafine particles

Despite growing evidence of their adverse health effects, the regulatory framework for UFPs remains limited, with no legally binding standards currently established at either the EU or global level. Cassee et al. (2019) reported that this is partly due to the lack of consistent epidemiological evidence on their independent adverse effects and identified several reasons for this, including: i) the absence of reference instrumentation and standardised methods for measuring PNC and PNSD; ii) the inconsistency in lower size detection limits (ranging from 3 to 20 nm) among studies, complicating comparisons

between UFP concentration datasets; iii) the limited availability of long-term exposure data; iii) the high spatial and temporal variability in UFP concentrations; iv) the limited representativeness due to the use of only a single monitoring site per city in most studies; and v) the diversity of sources contributing to UFP concentrations in different regions.

Although the WHO recognised that the current body of evidence is insufficient to formulate specific AQ guidelines for UFP, WHO (2021a) provided good practice recommendations aimed at addressing the monitoring and management of UFP exposure:

- Quantify ambient UFP in terms of PNC for a size range with a lower limit of ≤ 10 nm and no restriction on the upper limit.
- Integrate UFP monitoring into existing AQ monitoring, including size-segregated real-time PNC (PNSD) measurements.
- Distinguish between low and high PNC to guide decisions on the priorities of UFP source emission control. Low PNCs are defined as $< 1,000 \text{ \# cm}^{-3}$ (24-hour mean) and high PNCs as $> 10,000 \text{ \# cm}^{-3}$ (24-hour mean) or $20,000 \text{ \# cm}^{-3}$ (1-hour mean).
- Utilise emerging science and technology such as mobile sensors and real-time monitoring devices to the assessment UFP exposure for application in epidemiological studies and UFP management.

In the new Ambient Air Quality Directive (EU) 2024/2881 (EC, 2024), UFPs were recognised as pollutants of emerging concern and introduced for the first time in the legislation. While the directive does not yet establish legally binding limit of target values for UFPs, it mandates the monitoring of PNC and PNSD in urban supersites. Measurements are required to cover particles with a lower size limit of 10 nm and no restriction on the upper size limit. Additionally, the directive requires UFP monitoring at pollution hotspots such as traffic-heavy zones, airports, harbours, and industrial locations, as well as at rural supersites. Member States must establish at least one urban background supersite (with PNSD measurements) per ten million inhabitants, and one rural supersite per 100,000 km² in rural background areas. Furthermore, total UFP shall be measured, with one monitoring site per five million inhabitants, in areas prone to high UFP concentrations. Full implementation is targeted by 2030, with reviews every five years to

ensure that standards remain aligned with the latest scientific evidence and health guidelines.

By including UFPs in the monitoring network, the directive aims to enhance scientific understanding of their spatial and temporal variability, sources, and health impacts, thereby supporting improved AQ management and the development of health-based regulations across the EU.

1.4 Research gaps and the RI-URBANS context

Despite recent regulatory advances concerning UFPs and their recognition as pollutants of emerging concern, significant gaps remain in understanding their characteristics, sources, behaviours, and health impacts. The lack of comprehensive, normalised and standardised data on PNCs and PNSDs limits comparability across sites and studies and makes it difficult to assess their distribution and trends. Additionally, the absence of systematic measurements near hotspots, as required by the EC (2024), further hinders the identification of emerging sources and their impacts. Moreover, global urban UFPs source contributions are scarcely evaluated, complicating efforts to identify which emissions should be reduced and to what extent. Without SA, the relative impact of different emission sectors on both health and climate cannot be effectively evaluated. In addition, very few studies offer comparative long-term assessments across multiple locations, preventing a broader understanding of spatial and temporal variability. Accordingly, global studies on urban trends of UFP are not available. Addressing these gaps is essential to support evidence-based AQ management and targeted regulatory action.

The RI-URBANS project (Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial Areas) is a European research initiative funded by the EU Horizon 2020 programme (grant agreement No. 101036245). Its primary goal is to develop tools for the measurement and analysis of advanced AQ parameters in support of urban AQ monitoring, health impact assessment, and policy-making. Among its key objectives is the integration of UFP-PNSD into AQ assessments.

Within the framework of RI-URBANS, this PhD thesis focuses on characterising the spatial and temporal variability of UFPs in European urban environments, with particular emphasis on understanding their sources through long-term data analysis and SA techniques.

2 OBJECTIVES AND STRUCTURE

2.1 Objectives

The general objective of this thesis is to improve the understanding of the spatial and temporal variability and sources of UFPs in urban Europe. The enhanced understanding will contribute to: i) assessing health and climate impacts, ii) providing practical recommendations for UFP monitoring, and iii) devising targeted mitigation strategies to improve AQ.

To achieve this goal, the thesis addresses the following specific objectives:

O1. Compile long-term PNC and PNSD measurement data from (mostly) urban monitoring sites involved in the RI-URBANS project, as well as from additional non-partner sites across Europe, to ensure broad spatial coverage and enable comparative analysis across diverse urban environments.

O2. Treat and harmonise the collected PNSD datasets to facilitate comparability across monitoring sites, including the identification and removal of measurement errors, and the adjustment of particle size ranges to produce consistent and comparable PNSDs.

O3. Evaluate the spatial variability of PNCs and PNSDs across urban Europe, aiming to identify key similarities and major differences between site types (traffic, urban background, suburban background, and regional background) and climate zones, and to assess relationships with co-pollutants and meteorological parameters.

O4. Assess long-term trends in UFP concentrations and particle size modes across urban Europe, alongside co-pollutant concentrations and meteorological parameters to support the interpretation of observed temporal patterns. Understanding how UFPs have responded to past and current policies is essential to assess further abatement policies.

O5. Identify and quantify the sources contributing to PNSDs in urban environments across Europe using receptor modelling techniques, and evaluate long-term trends in these source contributions.

O6. Characterise PNSD profiles near a major European airport and quantify source contributions in areas influenced by aviation emissions, given that aviation is a relevant source of UFPs which is often mixed with nucleation sources and not distinctly identified in specific SA studies.

O7. Evaluate the differences in source contributions to UFPs using receptor modelling techniques, comparing results with and without dispersion normalisation and distinguishing between warm and cold periods through a comparative analysis based on long-term data from a European urban background site, in order to assess the impact of meteorology-driven dispersion on SA.

2.2 Structure of the thesis

This doctoral thesis is presented as a compendium of four scientific articles published in peer-reviewed journals and an additional one under review. The thesis is structured as follows:

Chapter 1 provides a comprehensive introduction to ultrafine particles, covering their size distributions, sources, and relevant atmospheric processes influencing their concentrations, as well as their impact on AQ, human health, and climate. It outlines the current state of research, highlights existing knowledge gaps, and presents the broader context of AQ standards and the RI-URBANS project to which this doctoral thesis is linked.

Chapter 2 defines the objectives of the thesis and its overall structure, including a list of the articles that comprise the compendium.

Chapter 3 describes the methodology employed in this thesis, detailing the areas of the study, instrumentation, and the data treatment and analysis techniques used.

Chapter 4 presents the five scientific articles, either published or under review, that constitute the compendium:

- **Paper I:** Trechera, P., **Garcia-Marlès, M.**, Liu, X., Reche, C., Perez, N., Savadkoohi, M., Beddows, D., Salma, I., Vörösmarty, M., Casans, A., Casquero-Vera, J.A., Hueglin, C., Marchand, N., Chazeau, B., Gille, G., Kalkavouras, P., Mihalopoulos, N., Ondracek, J., Zíková, N., Niemi, J.V., Manninen, H.E., Green, D.C., Tremper, A.H., Norman, M., Vratolis, S., Eleftheriadis, K., Gomez-Moreno, F.J., Alonso-Blanco, E., Gerwig, H., Wiedensohler, A., Weinhold, K., Merkel, M., Bastian, S., Petit, J.E., Favez, O., Crumeyrolle, S., Ferlay, N., Dos Santos, S.M., Putaud, J.P., Timonen, H., Lampilahti, J., Asbach, C., Wolf, C., Kaminski, H., Altug, H., Hoffmann, B., Rich, D. Q., Pandolfi, M., Harrison, R.M., Hopke, P.K., Petäjä, T., Alastuey, A., Querol, X., 2023. Phenomenology of ultrafine particle concentrations and size distribution across urban Europe. *Environment International*, 172, 107744. <https://doi.org/10.1016/j.envint.2023.107744>
- **Paper II:** **Garcia-Marlès, M.**, Lara, R., Reche, C., Pérez, N., Tobías, A., Savadkoohi, M., Beddows, D., Salma, I., Vörösmarty, M., Weidinger, T., Hueglin, C., Mihalopoulos, N., Grivas, G., Kalkavouras, P., Ondráček, J., Zíková, N., Niemi, J.V., Manninen, H.E., Green, D.C., Tremper, A.H., Norman, M., Vratolis, S., Eleftheriadis, K., Gómez-Moreno, F.J., Alonso-Blanco, E., Wiedensohler, A., Weinhold, K., Merkel, M., Bastian, S., Hoffmann, B., Altug, H., Petit, J.E., Favez, O., Dos Santos, S.M., Putaud, J.P., Dinoi, A., Contini, D., Timonen, H., Lampilahti, J., Petäjä, T., Pandolfi, M., Hopke, P.K., Harrison, R.M., Alastuey, A., Querol, X., 2024. Inter-annual trends of ultrafine particles in urban Europe. *Environment International*, 185, 108510. <https://doi.org/10.1016/j.envint.2024.108510>
- **Paper III:** **Garcia-Marlès, M.**, Lara, R., Reche, C., Pérez, N., Tobías, A., Savadkoohi, M., Beddows, D., Salma, I., Vörösmarty, M., Weidinger, T., Hueglin, C., Mihalopoulos, N., Grivas, G., Kalkavouras, P., Ondráček, J., Zíková, N., Niemi, J.V., Manninen, H.E., Green, D.C., Tremper, A.H., Norman, M., Vratolis, S., Diapouli, E., Eleftheriadis, K., Gómez-Moreno, F.J., Alonso-Blanco, E., Wiedensohler, A., Weinhold, K., Merkel, M., Bastian, S., Hoffmann, B., Altug, H., Petit, J.E., Acharja, P., Favez, O., Dos Santos, S.M., Putaud, J.P., Dinoi, A., Contini, D., Casans, A., Casquero-Vera, J.A., Crumeyrolle, S., Bourrianne, E., Van Poppel, M., Dreesen, F.E., Harni, S., Timonen, H., Lampilahti, J., Petäjä, T., Pandolfi, M.,

Hopke, P.K., Harrison, R.M., Alastuey, A., Querol, X., 2024. Source apportionment of ultrafine particles in urban Europe. *Environment International*, 194, 109149. <https://doi.org/10.1016/j.envint.2024.109149>

- **Paper IV: Garcia-Marlès, M.**, Peters, J., Van Laer, J., Frijns, E., Berghmans, P., Staelens, J., Hofman, J., de Vos, T., Brasseur, O., Van Poppel, M., Alastuey, A., Querol, X., 2025. Spatiotemporal variation and source apportionment of ultrafine particles near Brussels Airport (Belgium). *Atmospheric Environment*, 359, 121375. <https://doi.org/10.1016/j.atmosenv.2025.121375>
- **Paper V: Garcia-Marlès, M.**, Reche, C., Pérez, N., Alastuey, A., Querol, X., 2025. Effect of dispersion normalisation on long-term source apportionment and trends of ultrafine particles in Barcelona (NE Spain). Submitted to *Atmospheric Environment* on 2 September 2025; under review as of October 2025.

Chapter 5 provides a joint discussion of the results obtained in the five studies.

Chapter 6 summarises the main conclusions of this thesis.

Chapter 7 offers recommendations for future research in this field to fill some of the gaps in current knowledge.

Chapter 8 includes the conference presentations and relevant reports directly related with the thesis, as well as additional peer-reviewed scientific articles indirectly related.

Chapter 9 lists all references cited throughout the thesis, excluding those cited exclusively within the compiled papers.

Appendix contains the supplementary material related to the scientific articles presented.

Each of the articles included in this thesis addresses at least one of the specific objectives previously presented, as illustrated in Figure 2.1.

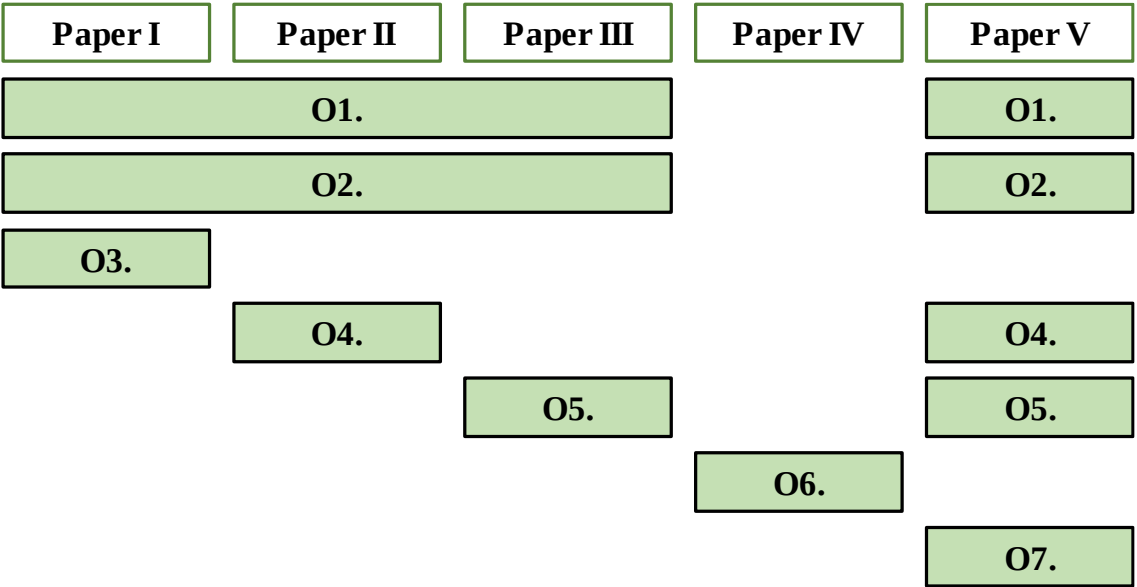


Figure 2.1: Schematic showing the integration of thesis objectives (O) within the scientific publications.

3 METHODOLOGY

3.1 Methodological approach

This chapter provides an overview of the study areas, the UFP-PNSD measurement instrumentation used to obtain the compiled datasets, and the methods employed for data processing and analysis. A more thorough description of the specific methodology used in each study can be found in its corresponding publication's methodology section. The overall methodological approach followed in this thesis is illustrated in Figure 3.1.

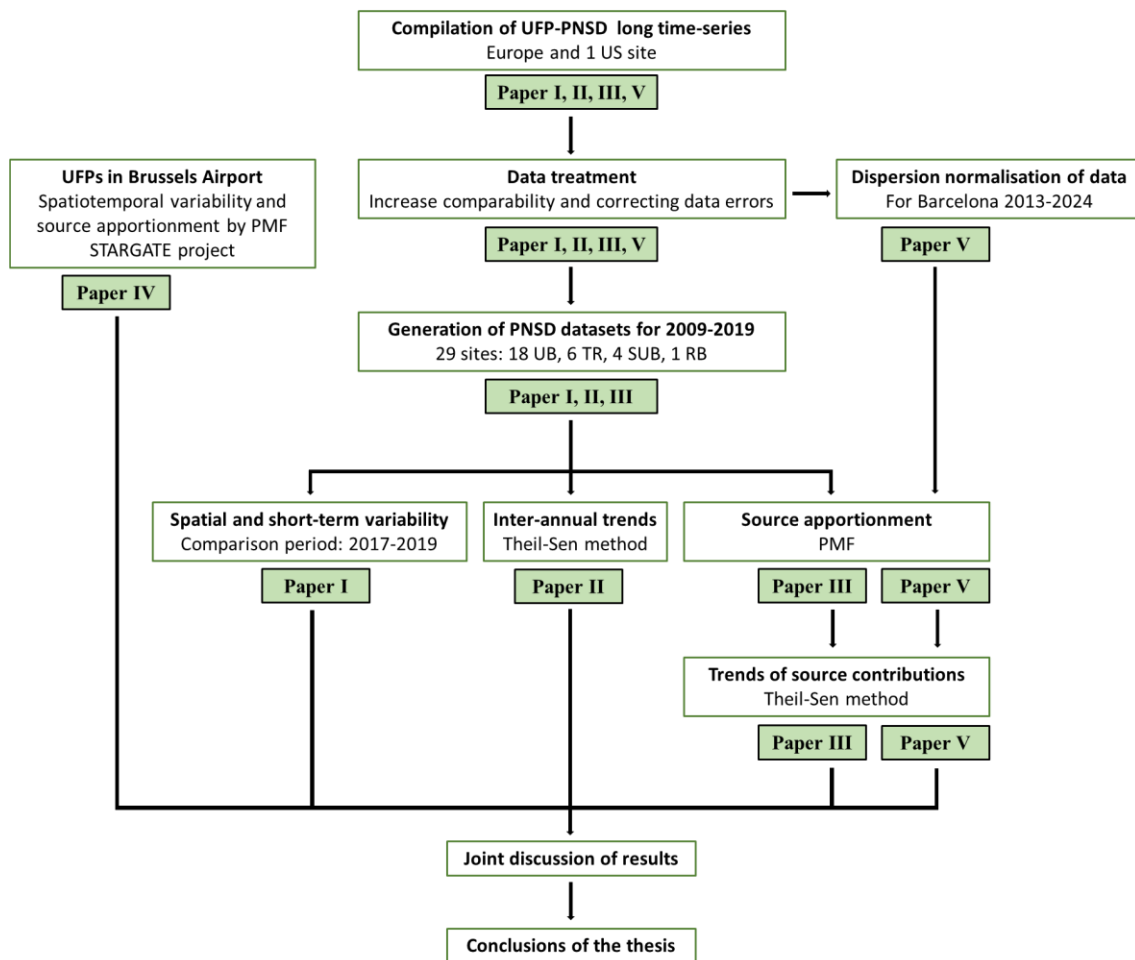


Figure 3.1: Schematic overview of the methodology approach used in this thesis, indicating the corresponding scientific publication for each step.

3.2 Measurement sites and instrumentation

3.2.1 Study sites and measurements

The articles included in this thesis are based on UFP-PNSD data collected from: i) AQ monitoring sites representing various background environments across Europe and one site in the United States (US), and ii) a monitoring campaign conducted in the vicinity of Brussels Airport.

Advanced AQ monitoring networks and research sites provided data covering twenty-nine locations across twenty-one cities in Europe and one in the US, through contributions from RI-URBANS partners and collaborators (Figure 3.2, Table 3.1). This broad range of sites enabled coverage of diverse climate zones, urban structures, PNCs, emission sources, and environmental types. The datasets included a total of eighteen urban background (UB), six traffic (TR), four suburban background (SUB), and one regional background (RB) sites. Each site was included in one or more of **Papers I, II, and III**, depending on data availability or other selection criteria, which are discussed in more detail later in this thesis. The BCN_UB site was also included in **Paper V**.

These measurements were carried out by the respective site data providers and shared for inclusion in the studies, with data typically provided as hourly averaged. In addition to PNSD, ancillary pollutant concentrations and meteorological parameters were collected when available, either from the same monitoring locations or a nearby AQ station. These included PM₁₀, PM_{2.5}, PM₁, BC, SO₂, NO₂, NO, O₃, and CO (Table 3.2), along with temperature (T), pressure (P), RH, WS, wind direction (WD), RAD, and rain (R).

Data were collected between 2009 and 2019 for **Papers I, II, and III**, depending on availability for each site (Table 3.2). This period was selected to provide relatively recent data while avoiding the influence of air pollution decreased due to the COVID-19 lockdown-associated measures, which were variable across urban Europe (Salma et al., 2020; Dinoi et al., 2021; Eleftheriadis et al., 2021; Petit et al., 2021; Putaud et al., 2021 and 2023). For **Paper I**, a common period of 2017–2019 was chosen to ensure a consistent time frame across all sites and to enhance inter-site comparability. For **Papers II and III**, a longer time span (2009–2019) was used to support trend and source analysis; sites with

less than three years of data were excluded, and some sites contributed partial records (e.g., 2013–2019), depending on data availability. For **Paper V**, which focused on the BCN_UB site, the dataset was extended to cover the period 2013–2024 to enable a robust long-term analysis. In this study, BLH data was also included.

All PNSD data used in **Papers I, II, III, and V** are openly available through the EBAS database infrastructure (<https://ebas.nilu.no/>).

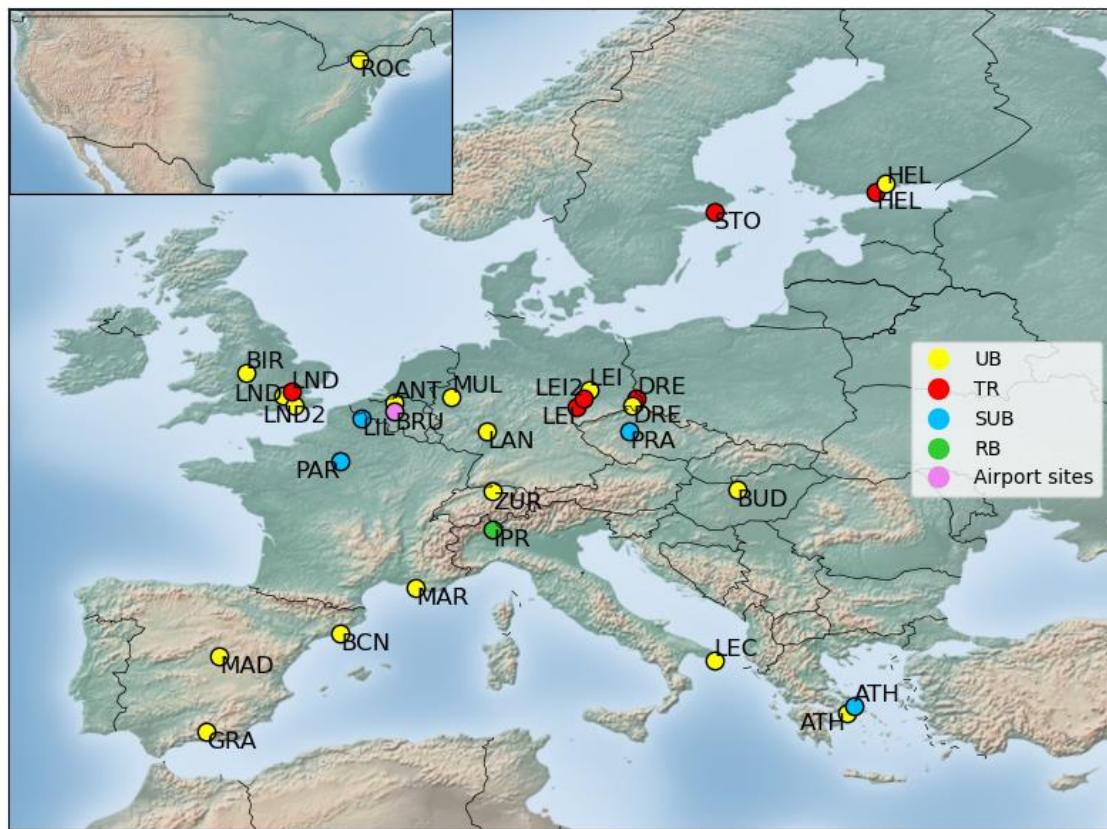


Figure 3.2: Map showing the location of the cities and type of station (UB, TR, SUB, and RB) supplying data on PNC and PNSD for **Papers I, II, III, and V**. Airport sites include four sites in the vicinity of Brussels Airport supplying data for **Paper IV**.

Table 3.1: List of sites providing UFP-PNSD datasets, including their site codes, coordinates, and the publication in which each site was used. Country codes follow the ISO 3166-1 alpha-2 standard. Coord., Coordinates; Alt., Altitude; a.s.l., above sea level.

City (Country)	Station name	Site code	Coord.; (Alt., m a.s.l.)	Paper
Antwerp (BE)	Borgerhout	ANT_UB	51.21 N, 4.43 E; (10)	III
Athens (GR)	Thissio	ATH_UB	38.00 N, 23.72 E; (110)	I, II, III
Barcelona (ES)	Palau Reial	BCN_UB	41.39 N, 2.13E; (80)	I, II, III, V
Birmingham (GB)	BAQS	BIR_UB	52.46 N, 1.93 W; (140)	I
Budapest (HU)	BpART Lab	BUD_UB	47.48 N, 19.06 E; (115)	I, II, III
Dresden (DE)	Winckelmann Str.	DRE_UB	51.04 N, 13.73 E; (120)	I, II, III
Granada (ES)	UGR	GRA_UB	37.16 N, 3.61 W; (680)	I, III
Helsinki (FI)	SMEAR III	HEL_UB	60.12 N, 24.58 E; (26)	I, II, III
Langen (DE)	UBA	LAN_UB	50.00 N, 8.39 E; (130)	I
Lecce (IT)	ECO Observatory	LEC_UB	40.20 N, 18.07 E; (50)	II, III
Leipzig (DE)	TROPOS	LEI_UB	51.35 N, 12.43 E; (113)	I, II, III
London (GB)	North Kensington	LND_UB	51.52 N, 0.21 W; (27)	I, II, III
London (GB)	Honor Oak Park	LND2_UB	51.45 N, 0.04 W; (36)	I
Madrid (ES)	CIEMAT-Moncloa	MAD_UB	40.45 N, 3.73 W; (669)	I, II, III
Marseille (FR)	Longchamp	MAR_UB	43.31 N, 5.39 E; (71)	I
Mülheim an der Ruhr (DE)	Mülheim-Styrum	MUL_UB	51.45 N, 6.87 E; (39)	I, II, III
Rochester NY (US)	NYS DEC	ROC_UB	43.15 N, 77.55 W; (137)	I, II, III
Zurich (CH)	Kaserne	ZUR_UB	47.38 N, 8.53 E; (410)	I, II, III
Dresden (DE)	North	DRE_TR	51.07 N, 13.74 E; (116)	I, II, III
Helsinki (FI)	Mäkeläkatu	HEL_TR	60.19 N, 24.95 E; (26)	I, II, III
Leipzig (DE)	Mitte	LEI_TR	51.34 N, 12.38 E; (111)	I, II, III
Leipzig (DE)	Eisenbahnstraße	LEI2_TR	51.35 N, 12.41 E; (120)	I
London (GB)	Marylebone Rd	LND_TR	51.52 N, 0.15 W; (35)	I, II, III
Stockholm (SE)	Hornsgatan	STO_TR	59.32 N, 18.05 E; (20)	I, II, III
Athens (GR)	Demokritos	ATH_SUB	37.99 N, 23.82 E; (270)	I, II, III
Lille (FR)	Villeneuve d'Ascq	LIL_SUB	50.61 N, 3.14 E; (70)	I, III
Paris (FR)	SIRTA	PAR_SUB	48.71 N, 2.16 E; (162)	I, II, III
Prague (CZ)	Suchdol	PRA_SUB	50.13 N, 14.38 E; (277)	I, II, III
Ispra (IT)	JRC	IPR_RB	45.80 N, 8.63 E; (209)	I, II, III
Monitoring campaign in the vicinity of Brussels Airport				
Zaventem (BE)	EE01	BRU Site A	50.88 N, 4.39 E; (24)	IV
Zaventem (BE)	MC03	BRU Site B	50.90 N, 4.46 E; (32)	IV
Zaventem (BE)	SZ04	BRU Site C	50.91 N, 4.51 E; (29)	IV
Zaventem (BE)	KM02	BRU Site D	50.94 N, 4.59 E; (14)	IV

Table 3.2: Measurement period and available ancillary pollutants of the sites providing UFP-PNSD for the studies of this thesis. ND, Not determined.

Site code	Period	Ancillary pollutants
ANT_UB	January 2017–December 2019	PM ₁₀ , PM _{2.5} , BC, SO ₂ , NO, NO ₂ , O ₃
ATH_UB	August 2015–December 2019	PM ₁₀ , BC, NO, NO ₂ , CO, O ₃
BCN_UB	January 2013–November 2024	PM ₁₀ , PM _{2.5} , PM ₁ , BC, SO ₂ , NO, NO ₂ , CO, O ₃
BIR_UB	July–December 2019	PM ₁₀ , PM _{2.5} , PM ₁ , BC, NO, NO ₂ , O ₃
BUD_UB	January 2009–December 2019	PM ₁₀ , SO ₂ , NO, NO ₂ , CO, O ₃
DRE_UB	December 2010–December 2019	PM ₁₀ , BC, SO ₂ , NO, NO ₂ , O ₃
GRA_UB	January 2017–December 2019	PM ₁₀ , BC, SO ₂ , NO, NO ₂ , CO, O ₃
HEL_UB	January 2009–December 2019	PM ₁₀ , PM _{2.5} , BC, SO ₂ , NO, NO ₂ , CO, O ₃
LAN_UB	January 2017–December 2019	PM ₁₀ , PM _{2.5} , PM ₁ , SO ₂ , NO, NO ₂ , CO, O ₃
LEC_UB	January 2015–December 2019	BC, SO ₂ , NO, NO ₂ , O ₃
LEI_UB	January 2009–December 2019	ND
LND_UB	January 2009–November 2018	PM ₁₀ , PM _{2.5} , PM ₁ , BC, SO ₂ , NO, NO ₂ , CO, O ₃
LND2_UB	November 2018–December 2019	PM ₁₀ , PM _{2.5} , PM ₁ , BC, NO, NO ₂ , O ₃
MAD_UB	July 2009–December 2019	PM ₁₀ , PM _{2.5} , BC, SO ₂ , NO, NO ₂ , CO, O ₃
MAR_UB	November 2018–May 2019	PM ₁₀ , PM _{2.5} , PM ₁ , BC, SO ₂ , NO, NO ₂ , O ₃
MUL_UB	March 2009–December 2019	PM ₁₀ , PM ₁ , BC, NO, NO ₂
ROC_UB	January 2009–December 2019	PM _{2.5} , BC, SO ₂ , NO, NO ₂ , CO, O ₃
ZUR_UB	January 2015–November 2019	PM ₁₀ , PM _{2.5} , BC, SO ₂ , NO, NO ₂ , CO, O ₃
DRE_TR	January 2009–December 2019	PM ₁₀ , BC, NO, NO ₂ , O ₃
HEL_TR	February 2015–June 2019	PM ₁₀ , PM _{2.5} , BC, NO, NO ₂ , CO, O ₃
LEI_TR	July 2010–December 2019	PM ₁₀ , BC, SO ₂ , NO, NO ₂
LEI2_TR	January 2017–December 2019	BC
LND_TR	January 2010–December 2019	PM ₁₀ , PM _{2.5} , PM ₁ , BC, SO ₂ , NO, NO ₂ , CO, O ₃
STO_TR	October 2010–December 2019	PM ₁₀ , PM _{2.5} , PM ₁ , BC, NO, NO ₂ , CO, O ₃
ATH_SUB	January 2010–December 2019	PM ₁₀ , PM _{2.5} , BC, NO, NO ₂ , O ₃
LIL_SUB	July 2017–December 2019	PM ₁₀ , PM _{2.5} , BC, NO, NO ₂ , O ₃
PAR_SUB	August 2012–December 2019	PM ₁₀ , PM _{2.5} , PM ₁ , BC, NO, NO ₂ , CO, O ₃
PRA_SUB	April 2012–December 2019	PM ₁₀ , PM _{2.5} , SO ₂ , NO, NO ₂ , CO, O ₃
IPR_RB	July 2009–December 2019	PM ₁₀ , BC, SO ₂ , NO, NO ₂ , CO, O ₃
Monitoring campaign in the vicinity of Brussels Airport		
BRU Site A	October–November 2015	PM ₁₀ , BC, NO _x
BRU Site B	October–November 2015	PM ₁₀ , BC, NO _x
BRU Site C	October–November 2015	PM ₁₀ , BC, NO _x
BRU Site D	October–November 2015	PM ₁₀ , BC, NO _x

PNSD data for **Paper IV** were obtained from a monitoring campaign conducted in the vicinity of Brussels Airport (Belgium) during October and November 2015 (Figure 3.2, Table 3.1, Table 3.2). The measurements were carried out by the Flanders Environment Agency (VMM) as part of the STARGATE project (funded by the EU's Horizon 2020 research and innovation programme, grant agreement GA101037053) and were subsequently made available for this study. The campaign included four monitoring sites (referred to as Sites A, B, C, and D), strategically located at varying distances from the airport along a transect roughly parallel to the main runway (25R/07L). Under prevailing southwesterly wind conditions, sites A and B were positioned upwind of the airport (at approximately 5 km and 250 m, respectively), while sites C and D were downwind (approximately 750 m and 7 km, respectively) (Figure 3.3). This setup enabled a detailed assessment of upwind and downwind effects under different wind scenarios, providing insights into dispersion patterns and the relative impact of aircraft activities. In addition to PNSD, mass concentration of ancillary pollutants (PM_{10} , BC, and NO_x) and meteorological parameters (T, RH, WS, WD, and R) were also collected.

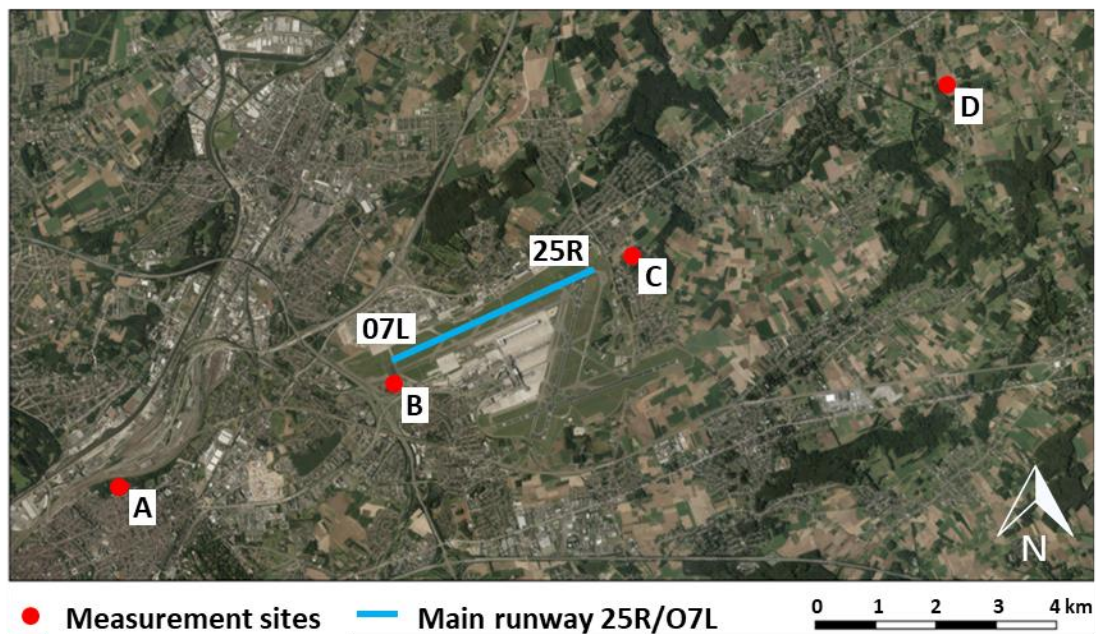


Figure 3.3: Measurement setup for **Paper IV**, showing the four monitoring sites (A, B, C, and D) located at varying distance from the main active runway 25R/07L of Brussels Airport.

3.2.2 Instrumentation

Throughout this thesis, data were collected using instruments deployed at long-term monitoring supersites (for **Papers I, II, III, and V**) and during a short-term measurement campaign (for **Paper IV**). The instrumentation used for PNSD measurements is summarised in Table 3.3, along with their respective particle size measurement ranges.

PNSD measurements were performed using Mobility Particle Size Spectrometers (MPSS), most commonly operated as Scanning Mobility Particle Sizer (SMPS) systems (Figure 3.4). These instruments are designed to measure the PNSD of submicron particles based on their electrical mobility (Wiedensohler et al., 2017). An SMPS typically consists of three main components: an aerosol neutraliser, an electrostatic classifier containing a Differential Mobility Analyser (DMA), and a Condensation Particle Counter (CPC). First, particles are neutralised to establish a predictable charge distribution before. Then they enter the DMA, where they are classified by electrical mobility (a property related to particle size) through the application of an electric field. A central negatively charged rod within the DMA attracts positively charged particles, allowing only those within a specific mobility range to pass through sample slit, while the remaining particles are discarded. These classified particles are then detected by the CPC, which enlarges them through condensation using butanol or water and counts them optically. By scanning across a range of voltages, the DMA sequentially selects particles of different sizes, allowing the system to build a complete particle size distribution over the defined diameter range during the measurement period.

In addition to SMPS instruments, similar systems were also used at some sites (Table 3.3), including the Differential Mobility Particle Sizer (DMPS), Twin Scanning Mobility Particle Sizer (TSMPS), Twin Differential Mobility Particle Sizer (TDMPS), and Optical Particle Counter (OPC). The DMPS was first developed by coupling the DMA to a continuous-flow CPC (Kangasluoma et al., 2020, and references therein). Later, Wang and Flagan (1990) improved the time resolution of the system to a few minutes by continuously scanning the DMA voltage and counting the particles in time bins, resulting in the SMPS. The TSMPS and TDMPS combine two DMA-CPC systems to cover a broader particle size range, typically from 3 to 800 nm.

Table 3.3: Instruments used to measure UFP-PNSD and their respective instrumental size ranges for the sites contributing data to the studies presented in this thesis. Grimm Aerosol Technik (GRIMM), TSI Incorporated (TSI), Leibniz Institute for Tropospheric Research (TROPOS), University of Helsinki (UHEL), and Stockholm University (SU) are the manufacturers or affiliated research institutions of the instruments.

Site code	UFP-PNSD Equipment	Range (nm)
ANT_UB	SMPS GRIMM C5420 long DMA	10–808
ATH_UB	SMPS TSI 3034	10–467
BCN_UB	SMPS TSI 3080 + CPC TSI 3772 and SMPS TSI 3082 + CPC TSI 3750	11–478
BIR_UB	SMPS TSI 3082 + CPC TSI 3750	12–552
BUD_UB	DMPS + CPC TSI 3775	6–994
DRE_UB	TSMPS TROPOS Vienna-type DMA 28 cm + CPC TSI 3010/3772	10–800
GRA_UB	SMPS TSI 3082 + CPC TSI 3772	11–496
HEL_UB	TDMPs Hauke-type DMA 10.9 cm + CPC TSI 3025/3756 and TDMPs Hauke-type DMA 28 cm + CPC TSI 3010/3772	3–891
LAN_UB	SMPS TSI 3080 + TSI 3772, rebuild at TROPOS incl. software	10–544
LEC_UB	SMPS TROPOS Vienna-type DMA 28 cm + CPC TSI 3772	10–800
LEI_UB	TDMPs TROPOS Vienna-type DMA 11 cm + CPC TSI 3025/3010	10–800
LND_UB	SMPS TSI 3080 + CPC TSI 3775 with long DMA	17–604
LND2_UB	SMPS TSI 3080 + CPC TSI 3775 with long DMA	17–604
MAD_UB	SMPS TSI 3080L + CPC TSI 3775	10–661
MAR_UB	SMPS TSI long DMA 3081A + CPC TSI 3752	15–661
MUL_UB	SMPS TSI 3080 + CPC TSI 3772	14–496
ROC_UB	SMPS TSI 3071 + CPC TSI 3010	11–470
ZUR_UB	SMPS TSI 3034 + Nafion aerosol dryer	11–478
DRE_TR	SMPS TROPOS Vienna-type DMA 28 cm + CPC TSI 3035/3010	5–800
HEL_TR	DMPS UHEL Vienna-type DMA + CPC Airmodus A20	6–800
LEI_TR	TDMPs TROPOS Vienna-type DMA 11 cm + CPC TSI 3025/3010	10–800
LEI2_TR	TDMPs TROPOS Vienna-type DMA 11 cm + CPC TSI 3025/3010	10–800
LND_TR	SMPS TSI 3080 + CPC TSI 3775 with long DMA	17–604
STO_TR	DMPS SU handmade + CPC TSI 3010/3775	10–410
ATH_SUB	SMPS TSI 3080 + CPC TSI 3772	10–550
LIL_SUB	SMPS TSI 3082 + CPC TSI 3750	16–594
PAR_SUB	SMPS GRIMM 5416 + OPC GRIMM	11–792
PRA_SUB	SMPS TSI 3034 rebuilt at TROPOS	9–519
IPR_RB	DMPS Vienna-type, homemade + CPC TSI 3010/3772	10–800
Monitoring campaign in the vicinity of Brussels Airport		
BRU Site A	SMPS TROPOS customised + CPC3772	9–848
BRU Site B	SMPS TROPOS customised + CPC3772	9–848
BRU Site C	SMPS TSI 3936L76 long DMA 3081 + TSI CPC 3776	8–284
BRU Site D	SMPS TSI 3936L76 long DMA 3081 + TSI CPC 3776	7–300

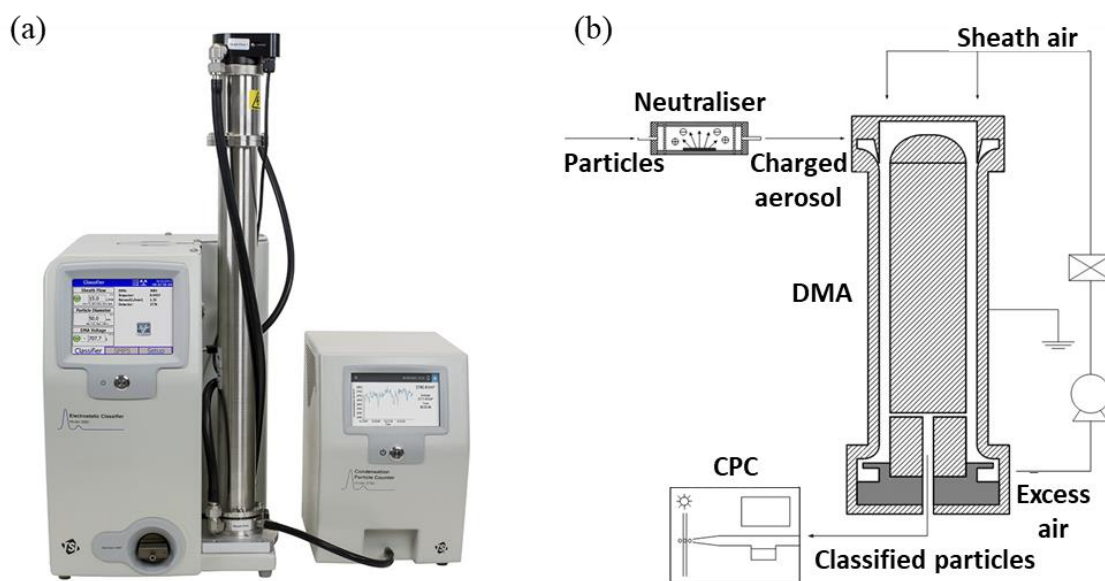


Figure 3.4: (a) SMPS model 3983, TSI. (b) Schematic of the SMPS system, adapted from Kim et al. (2009).

The specific size ranges and detection limits of these instruments vary depending on the model, configuration, and manufacturer. As a result, each measurement site reported PNSD data over slightly different size intervals (Table 3.3), although for this thesis the >10 nm size ranges were used in each case, to improve comparability.

In addition to PNSD measurements, a variety of instruments were used across the sites to monitor ancillary pollutants and meteorological parameters, either at the same locations or at nearby AQ monitoring stations, using instrumentation compliant with European standards:

- **BC**: Measured using the THERMO 5012 Multi-Angle Absorption Photometer (MAAP) and various Magee Scientific Aethalometers (AE), most commonly the AE33 model. These instruments estimate BC concentrations by detecting the light absorbed by aerosol particles collected on a filter (Petzold and Schönlinner, 2004).
- **PM**: Measured using instruments such as OPC, which detect light scattered by particles passing through a laser beam, allowing them to be sized and counted in real time. Other instruments to measure PM include Tapered Element Oscillating

Microbalance (TEOM), Beta Attenuation Monitoring (BAM), and Aerodynamic Particle Sizer (APS).

- Gaseous pollutants: Measured using automated analysers based on standardised techniques such as chemiluminescence (for NO_x), ultraviolet (UV) photometry (for O₃), UV fluorescence (for SO₂), and infrared absorption (for CO₂).
- Meteorological parameters: Measured using standard sensors for T, P, RH, WS, WD, RAD, and R. BLH data for **Paper V** were retrieved from the ERA5 atmospheric model (Hersbach et al., 2023), the fifth generation European Centre for Medium-range Weather Forecasts (ECMWF) reanalysis for the global climate and weather from 1940 onwards, produced by the Copernicus Climate Change Service (C3S).

3.3 Data treatment and harmonisation

Given the variation in size ranges and detection limits across the different instruments used at each site, the raw PNSD data required careful treatment and harmonisation before analysis.

Initially, data quality checks were performed to detect and remove errors or invalid measurements. In some datasets (BCN_UB, LIL_SUB, MAR_UB, PAR_SUB, and ZUR_UB), abrupt concentrations spikes in the smallest size bins were observed (Figure 3.5). These were likely caused by a combination of actual high concentrations due to localised plumes affecting these sizes, and measurement artefacts associated with low detection efficiency of the CPC at the lower end of the size distribution, as reported by Wiedensohler et al. (2012). Thus, the concentration values of the size bins lower than 12 nm in BCN_UB, 20 nm in LIL_SUB, 15 nm in MAR_UB, 11 nm in PAR_SUB, and 17 nm in ZUR_UB were deleted.

Additionally, harmonisation procedures were applied to standardised the datasets, accounting for differences in size detection limits to ensure comparability between sites and consistency in subsequent analyses. The minimum detection sizes across the twenty-nine sites included in **Papers I, II, III, and V** ranged from 3 to 20 nm, while the maximum detection sizes ranged from 410 to 994 nm (Figure 3.6). The upper detection limit has a

minor influence on total PNC, but the lower detection limit significantly affects concentrations in the nucleation mode. To harmonise the data and in line with established recommendations, size bins below 10 nm and above 800 nm were excluded from the datasets. It should be noted, after this process, not all sites retained the full 10–800 nm size range. These differences were duly considered when comparing PNC values across sites.

In **Paper IV**, data was also treated and harmonised. Lower size bins were excluded to minimise measurement errors, and a common size range from 10 to 280 nm was selected for the analysis.

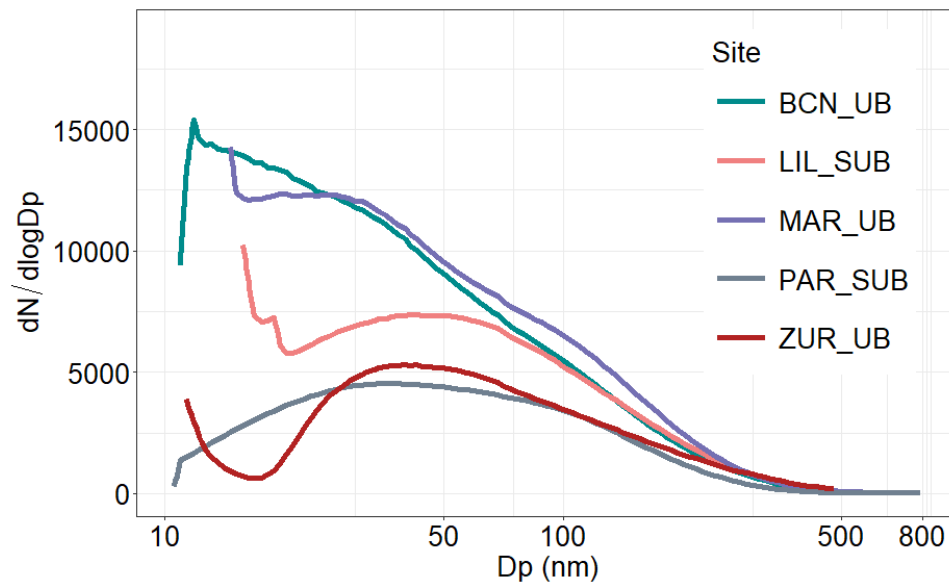


Figure 3.5: PNSDs for BCN_UB, LIL_SUB, MAR_UB, PAR_SUB, and ZUR_UB showing abrupt concentration spikes in the smallest size bins.

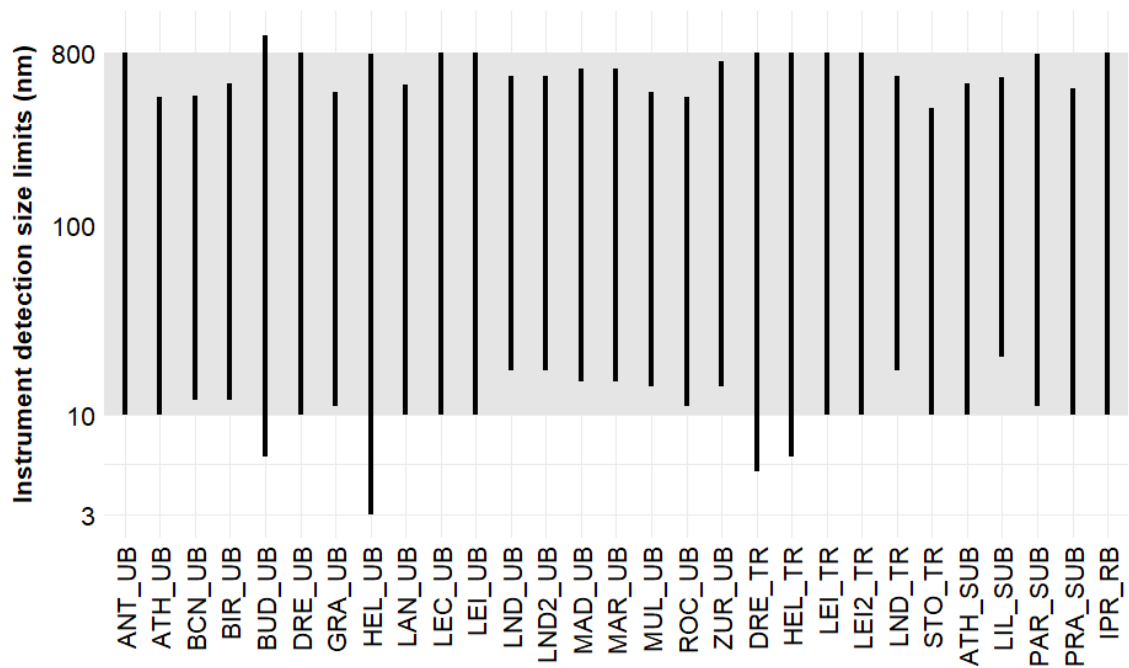


Figure 3.6: Particle size detection limits of PNSD measurements in **Papers I, II, III, and V** (black bars), and recommended harmonised range of 10–800 nm (grey).

3.4 Data analysis

Data treatment and statistical analysis were performed using R statistical software (v4.4.2, R Core Team, 2024) and the Openair package (Carslaw and Ropkins, 2012). The following sections detail the methodologies applied for trend analysis (used in **Papers II, III, and V**), SA (used in **Papers III, IV, and V**), and dispersion normalisation (used in **Paper V**).

3.4.1 Trend analysis

Data series were evaluated for monotonic trends over time using the non-parametric Theil-Sen method (Sen, 1968; Theil, 1992), implemented via the Openair package in R (Carslaw and Ropkins, 2012). This method, which is robust to outliers, is widely used in AQ trend assessments. The analysis was applied in **Papers II and III** at sites with datasets covering at least four complete years within a period exceeding five years, and in **Paper V** for the entire 12-year study period.

Monthly aggregation of the data was performed using a 30% threshold for hourly data availability. This threshold was chosen to balance data representativeness and coverage, considering the limited data availability (<50% at many sites) due to the complexity and maintenance requirements of UFP-PNSD measurements. Although other studies, such as Chen et al. (2022b), have used thresholds up to 40%, a 30% cut-off was found to be the most suitable after testing different thresholds.

Trend magnitude was quantified using the Theil-Sen slope, representing the annual change in concentration, along with associated confidence intervals and statistical significance (ss), providing insight into whether concentrations have increased, decreased, or remained stable over time.

For **Papers II** and **III**, a random effects meta-analysis was performed on the estimated trends to account for heterogeneity across sites. Individual slopes, expressed as percentage changes per year with their 95% confidence intervals, were combined to calculate mean effects for each site category (UB, TR, SUB, and RB) as well as overall, providing a comprehensive summary of the results. This analysis was conducted using the “meta” R package (Balduzzi et al., 2019).

Trend analyses were applied to hourly PNCs, different size modes, ancillary pollutants, and source contributions to PNCs, depending on the study. Table 3.4 summarises the variables analysed in each study.

Table 3.4: Overview of variables included in the trend analysis for each paper in this thesis. PNC refers to the total particle number concentration measured, while UFP refers to particles with diameters <100 nm.

Variable	Paper II	Paper III	Paper V
PNC	X		X
UFP	X		
Nucleation	X		X
Nucleation/PNC	X		
Aitken	X		X
Accumulation	X		X
PM ₁₀	X		X
PM _{2.5}	X		X
PM ₁	X		
SO ₂	X		X
NO	X		X
NO ₂	X		X
CO	X		X
O ₃	X		X
Source contributions to PNCs		X	X

3.4.2 Source apportionment

SA was conducted in **Papers III, IV, and V** to identify and quantify the sources of PNSD, using PMF. The growing use of PMF has been facilitated by the freely available implementation provided by the US Environmental Protection Agency (US EPA; Norris et al., 2014). However, the current US EPA PMF 5.0 program, a graphical user interface linked to the underlying solver, the Multilinear Engine 2 (ME-2; Paatero, 1999), is limited to datasets of approximately 500,000 data points. To address this limitation, Hopke et al. (2023) developed a tool enabling PMF analysis of large datasets using ME-2. This tool was employed in the studies presented in this thesis, as most datasets were long-term and exceeded the data point limit of the standard US EPA PMF interface.

For **Papers III and V**, hourly PNSD data were combined with hourly concentrations of ancillary pollutants available at each site, aiding in the identification of sources; each site was analysed separately. For **Paper IV**, the datasets had a time resolution of 5 minutes, and data from the four monitoring sites were combined so that PMF could resolve factors for all sites simultaneously.

PMF requires individual uncertainty estimates for each data point. The measurement uncertainties of PNSD were calculated using the methodology established by Ogulei et al. (2007) and further refined by Rivas et al. (2019), according to the following equation:

$$s_{ij} = \alpha_{ij} + C_3 \cdot n_{ij} \quad (2)$$

where:

$$\alpha_{ij} = 0.01(\bar{n}_{ij} + n_{ij}) \quad (3)$$

and n_{ij} is the observed concentration of sample i in size bin column j , $\bar{n}_{ij}$ is the arithmetic mean of the observed concentration in size bin j , and C_3 is a constant determined by trial-and-error, testing values between 0.001 and 0.15. Uncertainties for the 3% lowest and 3% highest size bins were multiplied by a factor of 2, and those for the subsequent 3% lowest and highest size bins by a factor of 1.5, to account for increased measurement errors reported in the lowest and highest size bins of the PNSD (Wiedensohler et al., 2017). Uncertainties for the ancillary pollutants were estimated using the equation provided by Polissar et al. (1998), and an additional uncertainty K was applied to all variables to optimise the model. The specific values of C_3 and K used in each analysis are reported in the corresponding papers.

In **Paper V**, the 12-year dataset comprised more than 6 million data points, exceeding the computational limit of the ME-2 tool developed by Hopke et al. (2023). Consequently, the dataset was divided into two subsets: a cold period (October, November, December, January, February, and March) and a warm period (April, May, June, July, August and September). This approach also enabled the investigation of seasonal variations in PNSD profiles, which are influenced by changes in atmospheric chemistry, as reported by Zhou et al. (2004) and Ogulei et al. (2007).

For each dataset, PMF was executed multiple times with different number of factors (sources) to determine the optimal solution. The best solutions were selected based on several diagnostic tools and criteria, following the recommendations of Belis et al. (2019) and Hopke et al. (2023), including: i) a Q/Q_{exp} ratio between 1 and 1.5, with values as close to 1 as possible; ii) scaled residuals approximately randomly distributed between -3 and 3, indicating a good fit; iii) assessment of factor displacement (DISP), which

evaluates the amount of rotational ambiguity in each preselected element of the factor profiles in response to small changes in the Q value; and iv) the interpretability of the resulting profiles and their temporal behaviour.

The objective function Q is the model parameter upon which PMF is optimised:

$$Q = \sum_i^n \sum_j^m \left(\frac{e_{ij}}{\alpha_{ij}} \right)^2 \quad (4)$$

where α_{ij} represents the uncertainty associated with each observation x_{ij} , and e_{ij} is the residual or error term. The model iteratively minimises Q until it falls below a convergence threshold, at which point the solution is considered optimised. The expected value Q_{exp} , or model degrees of freedom, is given by:

$$Q_{exp} = m \cdot n - p(m + n) \quad (5)$$

where m is the number of variables, n is the number of observations, and p is the number of factors.

Source identification was performed by analysing the PNSD profiles, daily and seasonal patterns, wind-related metrics (including WS and WD), and the contribution of each factor to the variance of the ancillary pollutants.

For **Papers III** and **V**, the trend analysis described earlier was applied to the source contributions to PNCs obtained from the PMF results.

3.4.3 Dispersion normalisation

Dispersion normalisation was applied to the data in **Paper V** to reduce the influence of meteorological variability on PNCs, using the ventilation coefficient (VC). The VC is an index that represents the volume of air available per unit time for the dilution of pollution released into the atmosphere (Ashrafi et al., 2009). For each measurement time period i , VC_i is defined as:

$$VC_i = BLH_i \times \bar{u}_i \quad (6)$$

where BLH_i is the boundary layer height during time period i and $\bar{u}_i$ is the mean WS. The measured concentration values (C_i , representing either PNC or mass concentrations) were then normalised by multiplying by the ratio of instantaneous VC_i to the average VC for the entire sampling period ($\overline{VC}$), resulting in the dispersion normalised concentration C_{vi} :

$$C_{vi} = C_i \times \frac{VC_i}{\overline{VC}} \quad (7)$$

After applying this method, SA was performed using the dispersion normalised data. The resulting source contributions were then unnormalised by dividing the resolved values by the same VC ratio to recover the actual (non-normalised) concentrations.

4 RESULTS

This chapter includes the five publications (four published and one under review) which conform the current thesis. Supplementary material is given in Appendix.

4.1 Summary of the publications

Paper I examines the PNSD profiles, as well as the levels and variability of UFPs during 2017–2019 across twenty-six sites in Europe and one in the US, covering TR, UB, SUB, and RB environments. PNCs progressively increased from Northern to Southern Europe, with substantially higher levels at TR sites. Daily patterns revealed typical traffic rush hours peaks, indicating road traffic as a major UFP source, while elevated midday PNCs at several sites suggested additional contributions from photochemical nucleation, fumigation, and shipping. To ensure reliable inter-site comparisons, PNSD data were first harmonised to a common particle size range of 10–800 nm, minimising artefacts arising from different lower size detection limits. In addition, analyses within the 25–800 nm range were performed to enable comparison across all sites with varying lower detection limits. This approach reduced discrepancies in total PNC and underscored the need for harmonised measurement protocols to ensure comparable UFP-PNSD datasets.

Paper II assesses long-term trends in PNC and particle size modes based on PNSD data from twenty-one sites, mostly those included in **Paper I**, covering the period 2009–2019. The Theil-Sen method and meta-analyses were conducted to evaluate overall and environment-specific trends. The results revealed a clear reduction in traffic-related emissions, with significant decreases in Aitken and accumulation mode particles, indicating the positive effects of road traffic policies implemented during the study period. In contrast, trends in the nucleation mode varied across cities, reflecting the absence of emission controls for this size range and the influence of additional sources such as photochemical nucleation, industry, shipping, and aircraft emissions.

Paper III presents SA results of PNSD data from twenty-four sites, mostly those included in **Paper I**, covering 2009–2019. Using receptor modelling with PMF, the study identified ten factors contributing to PNC: four traffic-related, photonucleation, urban background, domestic heating, two regional, and long-distance transport. Road traffic was the main contributor at all UB and TR sites, while photonucleation and domestic heating were also significant in many cities. Long-term trends in source contributions were also assessed, revealing a clear decrease in traffic-related UFP concentrations, whereas trends in photonucleation were more variable, influenced by reductions in the CS, which favoured NPF, or by decreases in UFP precursor emissions.

Paper IV investigates the impact of airport operations at Brussels Airport on local AQ during a two-month monitoring campaign in autumn 2015. Four monitoring sites, strategically located at varying distances from the airport and along a transect parallel to the main runway, measured PNSD, and SA using PMF was conducted to identify the main contributors to PNC. Results showed strong correlations between UFP concentrations, meteorological conditions, and aircraft landing and takeoff operations, with the smallest particles dominating PNC near the airport. UFP levels decreased with distance but remained detectable several kilometres away, with taxiing and takeoff identified as the main contributors near the airport, while road traffic, biomass burning, and regional background were more important further away. These findings highlight the spatial persistence, operational sources, and size distribution of aircraft-related UFPs.

Paper V evaluates the effect of dispersion normalisation on UFP SA and long-term trends at the UB site in Barcelona during 2013–2024. PNSDs were normalised using the VC, derived from BLH and WS, and analysed with PMF. Seven factors were identified (three traffic-related, two photonucleation, and two regional), with road traffic as the dominant contributor. Dispersion normalisation reduced the estimated contribution of traffic sources while increasing the relative contribution of photonucleation, reflecting the influence of atmospheric mixing conditions. Trend analysis showed significant decreases in traffic-related factors, highlighting the effectiveness of emission control policies, while photonucleation showed no clear trend. These results demonstrate the importance of accounting for dispersion in UFP SA and long-term trend assessment.

4.2 Publications

4.2.1 Paper I

Phenomenology of ultrafine particle concentrations and size distribution across urban Europe

Trechera, P., **Garcia-Marlès, M.**, Liu, X., Reche, C., Perez, N., Savadkoohi, M., Beddows, D., Salma, I., Vörösmarty, M., Casans, A., Casquero-Vera, J.A., Hueglin, C., Marchand, N., Chazeau, B., Gille, G., Kalkavouras, P., Mihalopoulos, N., Ondracek, J., Zíková, N., Niemi, J.V., Manninen, H.E., Green, D.C., Tremper, A.H., Norman, M., Vratolis, S., Eleftheriadis, K., Gomez-Moreno, F.J., Alonso-Blanco, E., Gerwig, H., Wiedensohler, A., Weinhold, K., Merkel, M., Bastian, S., Petit, J.E., Favez, O., Crumeyrolle, S., Ferlay, N., Dos Santos, S.M., Putaud, J.P., Timonen, H., Lampilahti, J., Asbach, C., Wolf, C., Kaminski, H., Altug, H., Hoffmann, B., Rich, D. Q., Pandolfi, M., Harrison, R.M., Hopke, P.K., Petäjä, T., Alastuey, A., Querol, X.

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Quartile: Q1

For this paper, I contributed to conceptualisation, methodology, data collection, treatment, processing, and visualisation, and was responsible for writing the manuscript.



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Full length article

Phenomenology of ultrafine particle concentrations and size distribution across urban Europe

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ABSTRACT

The 2017–2019 hourly particle number size distributions (PNSD) from 26 sites in Europe and 1 in the US were evaluated focusing on 16 urban background (UB) and 6 traffic (TR) sites in the framework of Research Infrastructures services reinforcing air quality monitoring capacities in European **URBAN** & industrial areas (RI-URBANS) project. The main objective was to describe the phenomenology of urban ultrafine particles (UFP) in Europe with a significant air quality focus.

The varying lower size detection limits made it difficult to compare PN concentrations (PNC), particularly PN₁₀₋₂₅, from different cities. PNCs follow a TR > UB > Suburban (SUB) order. PNC and Black Carbon (BC) progressively increase from Northern Europe to Southern Europe and from Western to Eastern Europe. At the UB sites, typical traffic rush hour PNC peaks are evident, many also showing midday-morning PNC peaks anti-correlated with BC. These peaks result from increased PN₁₀₋₂₅, suggesting significant PNC contributions from nucleation, fumigation and shipping.

Site types to be identified by daily and seasonal PNC and BC patterns are: (i) PNC mainly driven by traffic emissions, with marked correlations with BC on different time scales; (ii) marked midday/morning PNC peaks and a seasonal anti-correlation with PNC/BC; (iii) both traffic peaks and midday peaks without marked seasonal patterns. Groups (ii) and (iii) included cities with high insolation.

PNC, especially PN₂₅₋₈₀₀, was positively correlated with BC, NO₂, CO and PM for several sites. The variable correlation of PNSD with different urban pollutants demonstrates that these do not reflect the variability of UFP in urban environments. Specific monitoring of PNSD is needed if nanoparticles and their associated health impacts are to be assessed. Implementation of the CEN-ACTRIS recommendations for PNSD measurements would provide comparable measurements, and measurements of <10 nm PNC are needed for full evaluation of the health effects of this size fraction.

1. Introduction

In spite of the important advances in the science of aerosols and air quality, important scientific and environmental challenges remain unsolved, especially those related to source apportionment of the specific components of atmospheric particulate matter (PM), atmospheric processes influencing aerosols, and the associated climate and health impacts.

One of the issues requiring scientific attention is the origin and variability of ultrafine particles (UFP) (Knibbs et al., 2011; Cassee et al., 2019; Rivas et al., 2021). UFPs have been defined by CEN/TC 264/WG 32 as particles sized <100 nm in diameter (“ISO - ISO/TS 80004-2:2015 - Nanotechnologies — Vocabulary — Part 2: Nano-objects”). PM standards for air quality refer to ambient air mass concentrations below a given aerodynamic diameter per unit volume of air ($\mu\text{g m}^{-3}$), while UFP concentrations are most commonly (but not exclusively) measured in terms of particle number concentration (PNC) per unit volume of air (particle number or $\# \text{ cm}^{-3}$). Usually, total PNC is driven by UFP since, in general, >80% of the PNC is accounted for by UFP (Baldauf et al., 2016; Sun et al., 2019). Therefore, in this study we will treat PNC and UFP as synonymous.

It is widely recognised that exposure to PM negatively impacts human health (WHO, 2013, 2021a). In 2016, ambient air pollution accounted for almost seven million premature deaths per year (WHO, 2016), as derived from the aggravation of cardiovascular and respiratory diseases and cancers. Several studies have also shown that UFP can deeply penetrate the respiratory system, thus causing respiratory and cardiovascular diseases in humans (Cassee et al., 2019; Donaldson and Tran, 2002; Kelly and Fussell, 2012; Oberdörster et al., 2005; Salma et al., 2015; Tobías et al., 2018; Valavanidis et al., 2008; Weichenthal et al., 2013). The smaller size of UFPs not only allows them to reach the deeper parts of the respiratory system, but a fraction of these, reaching the alveoli, translocate and reach the circulatory system, and from there can reach any organ in the body (Cassee et al., 2019; Kreyling et al., 2002; Peters et al., 2006, among others). Furthermore, there are evidences that UFP might reach the brain through the olfactory nerve

(Maher et al., 2016; Oberdörster et al., 2004).

The new World Health Organisation Air Quality Guidelines (WHO, 2021b) identify UFP concentration as a relevant air quality parameter and find that, although there is a body of evidence for the health effects of UFPs, results are still inconsistent. Cassee et al. (2019) and Rivas et al. (2021) reported that this inconsistency may be, at least in part, due to methodological differences in measurements, to the lack of representation of UFP human exposure to UFPs resulting from the use of only a single monitoring station per city in most studies, and the different sources contributing to UFP concentrations in other cities/regions. Although the WHO (2021b) does not provide guideline values, the monitoring of concentrations of UFPs and black carbon (BC) is recommended to allow a more accurate evaluation of their health effects.

Online measurements of UFP-PNSD (UFP – particle number size distribution) are complex and are usually performed using electrical mobility analysers (Flagan, 1998) in which the particles are charged and separated in sizes by the application of an electrical field, and counted with a condensation particle counter (CPC). This electrical field can be stepped or scanned, allowing sequential measurement of aerosol size distribution. The measurements yield datasets of PNC for multiple-size bins. The particle diameters covered by these systems range from 3 to 16 nm (as lower size detection limits) up to 400 to 1000 nm (as the upper size limit). Since neither total PNC nor UFP-PNSD are included in the current air quality standards, there are no reference methods for these measurements in the context of air quality monitoring. However, CEN has recently developed recommendations for UFP-PNSD measurements from 10 to 800 nm (“CEN/TS 17434:2020 - Ambient air - Determination of the particle number size distribution of atmospheric aerosol using a Mobility Particle Size Spectrometer (MPSS)”) and for total PNC measurements (“CEN/TS 16976:2016 - Ambient air - Determination of the particle number concentration of atmospheric aerosol”). Furthermore, ACTRIS (2021) based on Wiedensohler et al. (2012) provide a set of recommendations for the outdoor measurement of both UFP-PNSD and PNC which are close to the CEN standards, but allow for measurement of particles with diameters below 10 nm if required (e.g., Lehtipalo et al., 2022; Manninen et al., 2016).

The lack of harmonisation between these measurements (both in terms of instrumentation and measurement conditions) makes the direct comparison of data from different cities very difficult: a large difference in the lower detection limit might account for a large difference in total PNC or the Nucleation mode (<25 nm). These differences might also lead to inconsistent results when relating UFPs to health effects (Cassee et al., 2019; Rivas et al., 2021). The major differences occur for the PNC of the smaller particle sizes (<25 nm where high PNC and low mass concentrations occur), and may vary over time (i.e., at midday nucleation periods or traffic rush hours) and space (Azimi et al., 2014; Baldauf et al., 2016; Fazli et al., 2019; Gani et al., 2021; Sun et al., 2019; Tobías et al., 2018; Von Bismarck-Osten et al., 2013; Wehner et al., 2002). Thus, primary UFP emitted by traffic are drastically reduced after rush hours, and these are diluted very fast, decreasing concentrations within 10 s of meters from the kerb site (Montagne et al., 2015; Zhu et al., 2002a,b); while secondary UFP might have a smother time and spatial variability (Dall'Osto et al., 2013).

According to a recent review on UFP source apportionment based on UFP-PNSD measurements by Hopke et al. (2022), the typically reported sources of UFP include nucleation, several traffic sources (fresh to aged), domestic and residential heating, regional secondary inorganic aerosols (i.e. regional nitrate and sulphate), particles associated with oxidants as represented by O₃ (i.e. regional secondary organic and inorganic aerosols) and other sources (such as biomass burning, urban background sources, industrial emissions, mixed sources, dust and unknown sources). Shipping and aviation might also contribute to increasing UFP concentrations with a prevalence of the lowest mode (Diesch et al., 2013; Lorentz et al., 2019; Stacey et al., 2021). When focusing on European urban background and traffic sites, traffic UFP contributions dominate PNC, followed by nucleation or new particle formation (NPF), with NPF varying widely according to the climatic region (i.e., it is usually higher in the high-insolation areas; but not consistently) (Bousiotis et al., 2021; Brines et al., 2015; Harrison et al., 2011; Kerminen et al., 2018; Petäjä et al., 2007; Reche et al., 2011; Rivas et al., 2021; Salma et al., 2011; Sun et al., 2019). Focusing on NO_x, several studies agree that one of the main sources of PNC and NO_x are vehicle engines, suggesting that this could be one of the factors behind their strongly correlated (Beevers et al., 2012; Grundström et al., 2015; Johansson et al., 2007). Sánchez Jiménez et al. (2012) corroborated this evidence and also suggest that correlations with NO_x or NO₂ are higher in traffic sites than in urban background sites, because traffic sites are mainly influenced by car emissions and urban background sites may be influenced by common local sources, resulting in a decrease in correlation between PNC and NO_x or PNC and NO₂. In addition, Rivas et al. (2020) also evidenced several sources in the urban background at different European sites where traffic sources were associated with elevated NO₂ concentrations. Wolf et al. (2017) found high levels of correlation between PNC and NO_x concentrations in the city of Augsburg, influenced by common local sources but in this case pointing to traffic and industrial sources as main factors. These studies found that, in urban areas of Europe, the main source contribution arises from road traffic and increases ambient air UFP concentrations with major modes from 15 to 50 nm, depending on the ageing stage (Harrison et al., 2011; Wehner et al., 2002).

Other relevant source contributions include regional UFPs (usually with 80–100 nm modes, Sun et al., 2019) via NPF and growth (e.g., Kerminen et al., 2018). The contribution from NPF is characterised by an increase in UFPs with diameters below 20 nm and has been described as occurring in urban areas in two ways. The first is fast nucleation following the morning UFP peak (caused by high primary semi-volatile exhaust emissions) that nucleates as these dilute and cool (Harrison et al., 2011). The second is the midday or midday-morning UFP peak caused by photochemical NPF occurring with lower levels of pollution (low condensation sink); this may be followed by particle growth, which is attributed to regional (nucleation and growth) or urban (nucleation burst) origins (Hopke et al., 2022; Kerminen et al., 2018; Stanier et al.,

2004). UFP concentrations could even be twice as high during NPF events than they are during non-events (e.g., Bousiotis et al., 2021; Casquero-Vera et al., 2021; Németh and Salma, 2014; Thén and Salma, 2022). Recent studies carried out with complex instrumentation to analyse the ion clusters that yield NPF in urban areas found that H₂SO₄ (and probably NH₃ and/or amines) initiate nucleation, while the growth of the nucleated particles is driven by multicomponent condensation of the oxidation products of semi-volatile organic compound (SVOCs) (Bousiotis et al., 2021; Brean et al., 2020; Kulmala et al. 2014; Yao et al., 2018).

RI-URBANS is a European research project (Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial Areas, the European Union's Horizon 2020 research and innovation program, 101036245) is a European research project which demonstrates the applications of advanced air quality service tools in urban Europe to the assessment of policy decisions in order to better abate pollution and supply accurate information for health studies. The measurement of UFP-PNSD and its applications in air quality assessment is a major goal of RI-URBANS. In this context, this study aims to compile existing UFP-PNSD measurements in urban Europe and to evaluate these data according to (i) the instrumental and methodological approaches implemented; (ii) the comparison of urban concentrations across Europe; (iii) the identification of similarities and major differences; and (iv) the evaluation of relationships with other pollutants, such as BC, PM_x (PM₁₀, PM_{2.5}, PM₁), and gaseous pollutants (SO₂, NO, NO₂, O₃, CO), and with meteorological parameters. The results of the source UFP apportionment for the datasets reported here will be presented in a subsequent article.

2. Methodology

2.1. Cities and sites providing UFP-PNSD data

This study is based on UFP-PNSD data from 2017 to 2019 provided by air quality monitoring networks and research supersites to RI-URBANS. These include (Fig. 1 and Table 1):

- Sixteen urban background (UB) sites covering most of Europe: Athens (ATH), Barcelona (BCN), Birmingham (BIR), Budapest (BUD), Dresden (DRE), Granada (GRA), Helsinki (HEL), Langen (LAN), Leipzig (LEI), two in London (LND and LND2), Madrid (MAD), Marseille (MAR), Mülheim (MUL), and Zurich (ZUR), along with and Rochester (ROC) in New York state in the USA.
- Six traffic (TR) sites in Central (C), Western (W) and Northern (N) Europe: DRE, HEL, two in LEI (LEI and LEI2), LND and Stockholm (STO), but none in Southern (S) Europe or Eastern (E) Europe. A TR air quality station BUD providing data on ancillary pollutants is used for interpreting the nearby BUD_UB site, because these pollutants are not measured at BUD_UB.
- Four suburban background (SUB) sites: ATH, Lille (LIL), Paris (PAR) and Prague (PRA).
- One regional background (RB) site in the Po Valley in Italy (Ispra, ISP), which was included because no datasets could be obtained from urban areas in the Po Valley, a pollution hotspot in Europe where most of the PM pollution has a regional origin.

Thus, in total, twenty-seven supersites provided hourly 2017–2019 data on UFP-PNSD for this study. This period was selected to obtain relatively recent data without the effect of the decrease of pollution due to the COVID-19 lockdown-associated measures, which varied both across urban Europe (Eleftheriadis et al., 2021; Putaud et al., 2021; Salma et al., 2020) and globally (Torkmahalleh et al., 2021).

From the twenty-seven sites of the study, twelve have already reported data to the ACTRIS data Centre (EBAS, <https://actris.nilu.no/Data/Policy/>) (ATH SUB, GRA_UB, HEL_UB, ISP_RB, MAD_UB, PAR SUB, PRA SUB, the three sites from LEI and the two from DRE), but fifteen

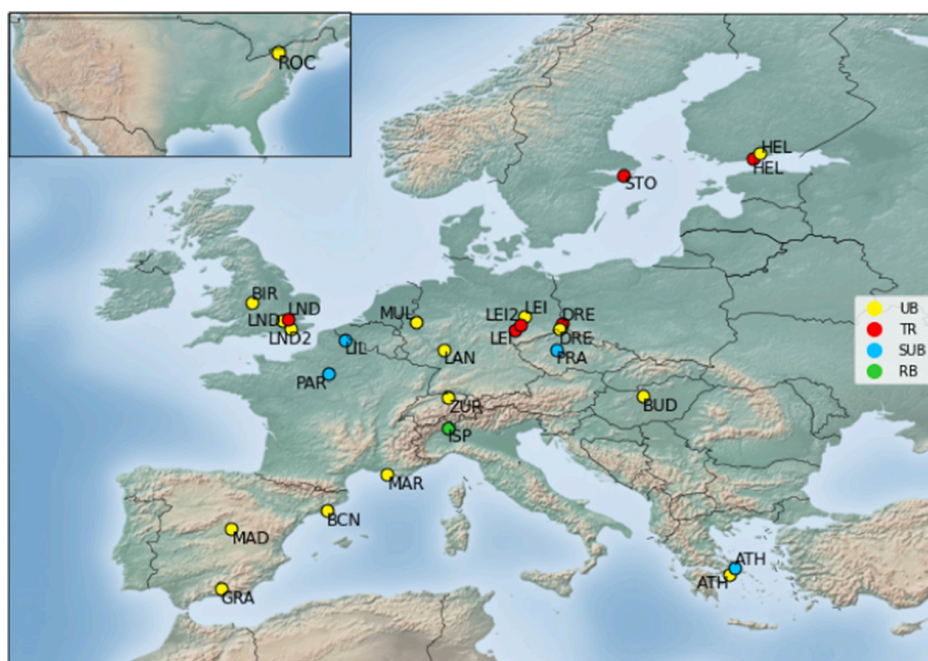


Fig. 1. Location of the cities supplying data on particle number concentrations and size distributions for the present study and the type of station. UB, Urban background; TR, Traffic; SUB, Suburban background; RB, Regional background.

stations have not. Data on co-measured pollutants and meteorological parameters were also collected, when available, from the same sites or nearby air quality monitoring sites.

The selection of cities allowed coverage of the entire European urban concentration ranges of urban pollutants other than UFP (NO , NO_2 , BC, PM_{10} , CO , SO_2 and O_3 , Table S1), and of meteorological patterns (Figure S1 and Table S1). The warmest cities were ATH and BCN (with average temperatures of 19°C), followed by MAD, MAR and GRA (17°C), BUD, LIL, ISP, ZUR, PAR, MUL, LEI, LAN and BIR ($14\text{--}12^\circ\text{C}$), LND, DRE, ROC, PRA and STO ($11\text{--}9^\circ\text{C}$) and HEL (7°C). Similarly, a wide range of insolation is also covered, with ATH, MAD, GRA and BCN at an average of $206\text{--}187\text{ W m}^{-2}$, followed by LIL, LAN, ZUR, STO, BUD and ISP ($179\text{--}150\text{ W m}^{-2}$) and PAR, HEL, DRE, LEI, BIR and MUL ($134\text{--}93\text{ W m}^{-2}$). The most humid cities were BIR, LIL, LND, LAN, PAR, HEL, MUL, PRA, BCN, ISP and DRE ($86\text{--}70\%$ average relative humidity), followed by STO, LEI, ROC, ZUR and BUD ($69\text{--}64\%$) and ATH, MAD and GRA ($56\text{--}48\%$). Average wind speed was higher in LND, PRA, STO and MAD ($4.3\text{--}3.4\text{ m s}^{-1}$), followed by HEL, MUL, ATH, PAR and BUD ($3.3\text{--}2.5\text{ m s}^{-1}$), LAN, ZUR, BCN, LEI, LIL and GRA ($2.2\text{--}1.4\text{ m s}^{-1}$) and DRE, MAR and ISP ($1.2\text{--}0.5\text{ m s}^{-1}$). However, the different altitudes at which the wind speed was measured in various cities makes it difficult to compare wind speeds appropriately.

2.2. Measurements

The instrumentation used for measuring UFP-PNSD and BC at the different stations is reported in Table 2. Meteorological parameters and concentrations of PM and gaseous pollutants were recorded in each (or nearest) station by instrumentation fulfilling European standards. It is important to note that the gaseous pollutants in Budapest were measured at the nearest station, which is characterised as a typical traffic station (BUD_TR). All the data received were averaged to hourly values.

Different instruments were used to obtain the compiled UFP-PNSD datasets; therefore, the measured size ranges measured varied. The instruments used were the Scanning Mobility Particle Sizer (SMPS), Differential Mobility Particle Sizer (DMPS), Twin Scanning Mobility

Particle Sizer (TSMPS) or Twin Differential Mobility Particle Sizer (TDMPS) (Table 2). ACTRIS and CEN guidance (see introduction) recommends a size measurement range of $10\text{--}800\text{ nm}$. However, lower sizes are measured for research purposes (specifically, to study nucleation episodes). Thus, some stations start measuring PNSD at finer sizes (HEL_UB and HEL_TR, 3 and 6 nm, respectively; DRE_TR, 5 nm; BUD_UB, 6 nm) or stop measuring PNSD at coarser upper limits (HEL_UB and TR, 891 nm; BUD_UB, 1000 nm). Nevertheless, the latter affects total PNC to a lower degree than the former because of the relatively low PNC within the coarser sizes. Furthermore, several sites start size measurements at 11 to 17 nm (BCN_UB, BIR_UB, GRA_UB, LIL_SUB, the three sites from LND, MAR_UB, MUL_UB, PAR_SUB, ROC_UB and ZUR_UB) rather than the recommended 10 nm, or stop measurements at lower sizes (410 to 792 nm, the two sites from ATH, BCN_UB, BIR_UB, GRA_UB, LAN_UB, MUL_UB, LIL_SUB, the three sites from LND, MAD_UB, MAR_UB, PAR_SUB, PRA_SUB, ROC_UB, STO_TR and ZUR_UB) than the recommended 800 nm.

2.3. Data treatment

Data treatment and statistical analysis were carried out using R statistical software (v4.1.3) and the *Openair* package (Carslaw and Ropkins, 2012).

3. Results and discussion

3.1. Concentrations of ancillary pollutants

Mean concentrations of NO_2 (Table S2) reached $80\text{ }\mu\text{g m}^{-3}$ in LND_TR and $46\text{ }\mu\text{g m}^{-3}$ in BUD_TR, $30\text{--}36\text{ }\mu\text{g m}^{-3}$ in the LEI_TR, STO_TR, GRA_UB, HEL_TR and LND_UB sites, $25\text{--}30\text{ }\mu\text{g m}^{-3}$ in the ATH_UB, MAR_UB, DRE_TR, BCN_UB, LND2_UB and LIL_SUB sites, $15\text{--}25\text{ }\mu\text{g m}^{-3}$ in the ZUR_UB, LAN_UB, MUL_UB, DRE_UB, BIR_UB, MAD_UB, PRA_SUB and ROC_UB sites, and lower than $15\text{ }\mu\text{g m}^{-3}$ in the ATH_SUB, PAR_SUB, HEL_UB and ISP_RB sites. NO concentrations followed a similar trend with slight differences. Thus, as expected, the highest NO_2 and NO concentrations mainly occurred at the $\text{TR} > \text{UB} > \text{SUB}$ and RB sites, as in

Table 1

List of air quality sites supplying UFP-PNSD datasets to this study with location and type of environment. Coor., Coordinates; Alt., Altitude; UB, Urban Background; TR, Traffic; SUB, Suburban Background; RG, Regional Background.

City (Country)	Station Name	Station type	Acronym	Coor.; (Alt., m a.s.L.)
Athens (GR)	Thissio	UB	ATH_UB	38.00 N, 23.72 E; (110)
Barcelona (ES)	Palau Reial	UB	BCN_UB	41.39 N, 2.13E; (80)
Birmingham (GB)	BAQS	UB	BIR_UB	52.46 N, 1.93 W; (140)
Budapest (HU)	CAAG	UB	BUD_UB	47.48 N, 19.06 E; (115)
Dresden (DE)	Winckelmannstraße	UB	DRE_UB	51.04 N, 13.73 E; (120)
Granada (ES)	UGR	UB	GRA_UB	31.18 N, 3.58 W; (680)
Helsinki (FI)	SMEARIII	UB	HEL_UB	60.12 N, 24.58 E; (26)
Langen (DE)	UBA	UB	LAN_UB	50.00 N, 8.39 E; (130)
Leipzig (DE)	TROPOS	UB	LEI_UB	51.35 N, 12.43 E; (113)
London (GB)	North Kensington	UB	LND_UB	51.52 N, 0.21 W; (27)
London (GB)	Honor Oak Park	UB	LND2_UB	51.45 N, 0.04 W; (36)
Madrid (ES)	CIEMAT-Moncloa	UB	MAD_UB	40.45 N, 3.73 W; (669)
Marseille (FR)	Longchamp	UB	MAR_UB	43.31 N, 5.39 E; (71)
Mülheim an der Ruhr (DE)	Mülheim-Styrum	UB	MUL_UB	51.45 N, 6.87 E; (39)
Rochester NY (US)	NYS DEC	UB	ROC_UB	43.15 N, 77.55 W; (137)
Zurich (CH)	Kaserne	UB	ZUR_UB	47.38 N, 8.53 E; (410)
Dresden (DE)	North	TR	DRE_TR	51.09 N, 13.76 E; (116)
Helsinki (FI)	Mäkeläinkatu Supersite	TR	HEL_TR	60.19 N, 24.95 E; (26)
Leipzig (DE)	Mitte	TR	LEI_TR	51.34 N, 12.38 E; (111)
Leipzig (DE)	Eisenbahnstraße	TR	LEI2_TR	51.35 N, 12.41 E; (120)
London (GB)	Marylebone Road	TR	LND_TR	51.52 N, 0.15 W; (35)
Stockholm (SE)	Hornsgatan	TR	STO_TR	59.32 N, 18.05 E; (20)
Budapest (HU)	OLM Széna Square	TR	BUD_TR	47.51 N, 19.03 E; (117)
Athens (GR)	Demokritos	SUB	ATH_SU	37.99 N, 23.82 E; (270)
Lille (FR)	Villeneuve d'Ascq	SUB	LIL_SUB	

Table 1 (continued)

City (Country)	Station Name	Station type	Acronym	Coor.; (Alt., m a.s.L.)
Paris (FR)	SIRTA	SUB	PAR_SUB	50.61 N, 3.14 E; (70)
Prague (CZ)	Schudol	SUB	PRA_SUB	48.71 N, 2.16 E; (162)
				50.13 N, 14.38 E; (277)
Ispra (IT)	JRC	RG	ISP_RB	45.80 N, 8.63 E; (209)

urban Europe traffic is the primary source of these pollutants (Nguyen et al., 2018). An increasing concentration trend was also evident from N to S Europe for UB sites, with BIR_UB, ROC_UB and HEL_UB recording the lowest concentrations and cities in E and S Europe the highest, although LND_UB was also among the highest.

Mean concentrations of CO reached the highest value (0.66 mg m^{-3}) in BUD_TR and 0.16 to 0.37 mg m^{-3} at all other sites (excluding the RB) (Table S2). Concentrations of this pollutant were not higher at TR sites compared with UB and SUB sites. Thus, sources beside traffic, such as domestic biomass or coal burning (Querol et al., 2021), were expected. However, problems in measuring this pollutant at low concentrations might also negatively affect comparisons between cities.

Average SO_2 concentrations were $<2.2 \text{ } \mu\text{g m}^{-3}$ in most cities, except for PRA_SUB, DRE_UB, MAD_UB, BUD_TR, LND_TR, GRA_UB and ATH_UB, where concentrations reached 2.6 to $7.0 \text{ } \mu\text{g m}^{-3}$ (Table S2). Higher concentrations are probably associated with the domestic use of coal or oil (EEA, 2021).

Average O_3 concentrations fell in the range of 40 – $60 \text{ } \mu\text{g m}^{-3}$ in most cities except for LND_TR, BUD_TR, MAD_UB, ATH_UB and ATH_SUB with 19 , 27 , 67 , 74 and $82 \text{ } \mu\text{g m}^{-3}$, respectively (Table S2). The traffic sites recorded relatively low concentrations due to the titration of O_3 by NO. Annual urban background O_3 concentrations were higher in HEL (N Europe) than in BCN (S Europe); this was the converse of what was occurring in the background concentrations (EEA, 2021). The higher urban NO concentrations observed over BCN are probably consuming O_3 , leading to low O_3 concentrations.

PM_{10} concentrations were markedly higher in the UB sites of BUD, ATH and GRA (33 – $37 \text{ } \mu\text{g m}^{-3}$), intermediate in ISP_RB and the TR sites of STO, LND, LEI and DRE, PRA_SUB and BCN-UB (20 – $25 \text{ } \mu\text{g m}^{-3}$), and lower in LIL_SUB, MUL_UB, DRE_UB, HEL_TR, MAR_UB, ATH_SUB, LND2_UB, MAD_UB, ZUR_UB, PAR_SUB, LND_UB, LAN_UB, BIR_UB and HEL_UB (10 – $19 \text{ } \mu\text{g m}^{-3}$). The highest average $\text{PM}_{2.5}$ concentrations were recorded in ATH_UB, BUD_UB and PRA_SUB (16 – $19 \text{ } \mu\text{g m}^{-3}$), while intermediate (11 – $15 \text{ } \mu\text{g m}^{-3}$) and lower (4.2 – $9.2 \text{ } \mu\text{g m}^{-3}$) concentrations were recorded in the same groups of sites listed above for PM_{10} , with slight variations. PM_1 concentrations were measured only in MUL_UB, LND_TR, LND2_UB, LAN_UB, MAR_UB, BCN_UB, LND_UB, BIR_UB, PAR_SUB and STO_TR (12 , 11 , 10 , 9.1 , 8.9 , 8.6 , 8.0 , 7.6 , 7.5 and $6.3 \text{ } \mu\text{g m}^{-3}$, respectively). The narrower concentration ranges of PM_1 compared with that of $\text{PM}_{2.5}$ probably reflects the higher impact of dust (road dust, desert dust or construction dust) on $\text{PM}_{2.5}$ concentrations in the drier MAR and BCN sites compared to the C European cities.

As expected, the twenty-four available datasets of twenty-seven sites indicate that BC concentrations were higher in the TR sites of LND, LEI and LEI2 (1.5 – $2.7 \text{ } \mu\text{g m}^{-3}$), as well as in the five S European UB sites at GRA, ATH, MAR, BCN and MAD and at ISP_RB (1.2 – $2.0 \text{ } \mu\text{g m}^{-3}$), and were intermediate in the UK and C European cities (LND2_UB, LIL_SUB, LEI_UB, MUL_UB, LND_UB, BIR_UB, ATH_SUB, DRE_UB and PAR_SUB) and the TR sites of DRE, STO and HEL (0.91 – $1.1 \text{ } \mu\text{g m}^{-3}$). The lowest concentrations were recorded in the UB sites of ZUR, ROC and HEL (0.33 – $0.48 \text{ } \mu\text{g m}^{-3}$). BUD, with high levels of traffic-related pollutants,

Table 2

Instruments used to measure UFP-PNSD and BC in the different stations. ND, Not-determined; SMPS, Scanning Mobility Particle Sizer; CPC, Condensation Particle Counter; MAAP, Multi-Angle Absorption Photometer; DMPS, Differential Mobility Particle Sizer; TSMPS, Twin Scanning Mobility Particle Sizer; DMA, Differential Mobility Analyser; TDMPS, Twin Differential Mobility Particle Sizer; UCPC, Ultrafine Condensation Particle Counter; OPC, Optical Particle Counter; UB, Urban Background; TR, Traffic; SUB, Suburban Background; RG, Regional Background.

Station Name	UFP-PNSD Equipment	Range (nm)	BC Equipment
ATH_UB	SMPS TSI 3080L + CPC TSI 3772	10–467	Magee Scientific Aethalometer AE33
BCN_UB	SMPS TSI 3080 + CPC TSI 3772	12–478	THERMO 5012 MAAP
BIR_UB	SMPS TSI 3082 + CPC TSI 3750	13–552	Magee Scientific Aethalometer AE33
BUD_UB	DMPS + CPC TSI 3775	11–816	ND
DRE_UB	TROPOS-TSMPS uses a Vienna-type DMA 28 cm + CPC TSI 3010/3772	10–800	THERMO 5012 MAAP
GRA_UB	SMPS TSI 3082 + CPC TSI 3772	11–496	THERMO 5012 MAAP
HEL_UB	TDMPS Hauke-type DMA 10.9 cm + CPC TSI 3025	10–794	THERMO 5012 MAAP
LAN_UB	SMPS TSI 3080 + TSI 3772, rebuild at TROPOS incl. software	10–544	ND
LEI_UB	TROPOS-TDMPS uses a Vienna-type DMA 11 cm + UCPC TSI 3025	10–800	THERMO 5012 MAAP
LND_UB	SMPS TSI 3080 + CPC TSI 3775 with long DMA	17–604	Magee Scientific Aethalometer AE33
LND2_UB	SMPS TSI 3080 + CPC TSI 3775 with long DMA	17–604	Magee Scientific Aethalometer AE33
MAD_UB	SMPS TSI 3080L + CPC TSI 3775	15–661	Magee Scientific Aethalometer AE33
MAR_UB	SMPS TSI long DMA 3081A + CPC TSI 3752	15–661	Magee Scientific Aethalometer AE33
MUL_UB	SMPS TSI 3080 + CPC TSI 3772	14–496	Magee Scientific Aethalometer AE33
ROC_UB	SMPS TSI 3071 + CPC TSI 3010	11–467	Magee Scientific Aethalometer AE21
ZUR_UB	SMPS TSI 3034 + Nafion aerosol dryer	17–478	Magee Scientific Aethalometer AE33
DRE_TR	TROPOS-SMPS uses a Vienna-type DMA 28 cm + CPC TSI 3772	10–800	THERMO 5012 MAAP
HEL_TR	UHEL DMPS Vienna-type DMA + CPC Airmodus A20	11–800	THERMO 5012 MAAP
LEI_TR	TROPOS-TDMPS uses a Vienna-type DMA 11 cm + UCPC TSI 3025	10–800	THERMO 5012 MAAP
LEI2_TR	TROPOS-TDMPS uses a Vienna-type DMA 11 cm + UCPC TSI 3025	10–800	THERMO 5012 MAAP
LND_TR	SMPS TSI 3080 + CPC TSI 3775 with long DMA	17–604	Magee Scientific Aethalometer AE33
STO_TR	SMPS TSI 3071 + CPC TSI 3775	10–410	Magee Scientific Aethalometer AE33
ATH_SUB	SMPS TSI 3080 + CPC TSI 3772	10–550	Magee Scientific Aethalometer AE33
LIL_SUB	SMPS TSI 3082 + CPC TSI 3750	20–594	Magee Scientific Aethalometer AE33
PAR_SUB	SMPS GRIMM 5416 + OPC GRIMM	11–792	Magee Scientific Aethalometer AE33
PRA_SUB	SMPS TSI 3034 rebuilt at TROPOS	10–519	ND
ISP_RB	DMPS Vienna-type, home-made + CPC TSI 3010/3772	10–800	THERMO 5012 MAAP

did not measure BC.

3.2. Particle number size distributions

The variability of UFP-PNSD concentrations over time is complex

due to dynamic processes such as meteorology, emissions and atmospheric processes such as NPF (nucleation), evaporation, condensation, coagulation and deposition (Bousiotis et al., 2021; Harrison et al., 2019; Qi et al., 2015).

As stated in the methodology, comparing concentrations in the compiled PNC datasets was difficult due to the different size detection limits used. Furthermore, in five datasets (BCN_UB, LIL_SUB, MAR_UB, PAR_SUB, ZUR_UB) drastic positive or negative concentrations jumps in the ending lower sizes were detected (Figure S2). This directly affects the UFP PNC and it is for this reason that it has been decided to remove these lower size ranges. This signal is most likely the result of a combination of real concentrations at these sizes and measurement challenges at the low end of size distributions. In the urban environment, size distributions are highly dynamic and variable over a time scale ranging from seconds to minutes. Size distributions are measured by sequentially scanning or stepping through the size range. Close to a source of ultrafine particles, such as in traffic environments, a very localised plume can consequently influence only one or a few size bins. Furthermore, at size ranges close to 10 nm data inversion will amplify small errors due to concurrent low detection efficiency of the CPC and challenges in setting small classification voltages precisely and consistently. This challenge is evident in the study by Wiedensohler et al. (2012) as a higher instrument-to-instrument variability in the size range below 20 nm. The next step to improve the data quality is utilisation of specific instruments for the smaller size ranges (Kangasluoma et al., 2020), which would be operated in parallel with the DMPS/SMPS systems. In this study, to circumvent these challenges for the measurement of UFP-PNSD, the concentration values of the size bins lower than 12 nm in BCN_UB, 20 nm in LIL_SUB, 15 nm in MAR_UB, 11 nm in PAR_SUB and 17 nm in ZUR_UB were deleted (see ‘corrected range’ in Table S1 and Figure S2). Thus, the minimum detection sizes at all the twenty-seven sites ranged from 3 to 20 nm and the maximum detection sizes ranged from 410 to 1000 nm. The coarser detection size has a small influence on the total PNC. However, the lower size detection limit has a considerable impact, especially on the Nucleation mode (N_{x-25}) concentrations. As an example, HEL_UB measured UFP-PNSD with a size coverage from 3 to 794 nm and Lille from 20 to 594 nm. If we consider the range of 10 to 794 nm or 20 to 794 nm for HEL_UB the total PNC is reduced by 14% or 35% with respect to the 3 to 794 nm measurement. The nucleation mode PNC is reduced by 33% from N_{3-25} to N_{10-25} and by 82% from N_{3-25} to N_{20-25} . However, for HEL_UB, the difference in the PNC from 3–794 nm and 3–594 nm is <0.1%, because the low PNC in the coarse range of the Accumulation mode.

However, only fifteen of the datasets had >70% data availability for the study period and seven even had data availability of <50%. This deficiency reflects the complexity of the UFP-PNSD measurements and the need for detailed supervision and frequent instrumentation maintenance. RI-URBANS strongly recommends not only following the CEN and ACTRIS protocols for measuring PNSD, but also following the guidelines provided by Wiedensohler et al. (2012 and 2018) for the harmonisation of technical standards and data structures to facilitate high quality long-term observations of atmospheric PNSDs and calibration procedures by Wiedensohler et al. (2012 and 2018).

This study analysed the N_{10-800} and N_{25-800} concentrations to reduce this effect. In any case, when comparing PNC, one should consider site-specific lower detection sizes. For instance, N_{20-594} (LIL_SUB) and N_{10-800} (HEL_UB) were compared as N_{10-800} = PNC, despite their different sizes.

Once the datasets were corrected and the N_{10-800} selected, Fig. 2 shows the UFP-PNSD data were classified into the following four groups (as shown Fig. 2):

TR sites (DRE, HEL, LEI, LEI2, STO, LND). These sites have generally high PNCs, with the highest values in the Nucleation mode due to freshly emitted and quickly nucleated particles from traffic exhaust emissions (Harrison et al., 2011; Rönkkö et al., 2017; Sun et al., 2019; Wehner et al., 2002; among others). LND presents low concentrations in the

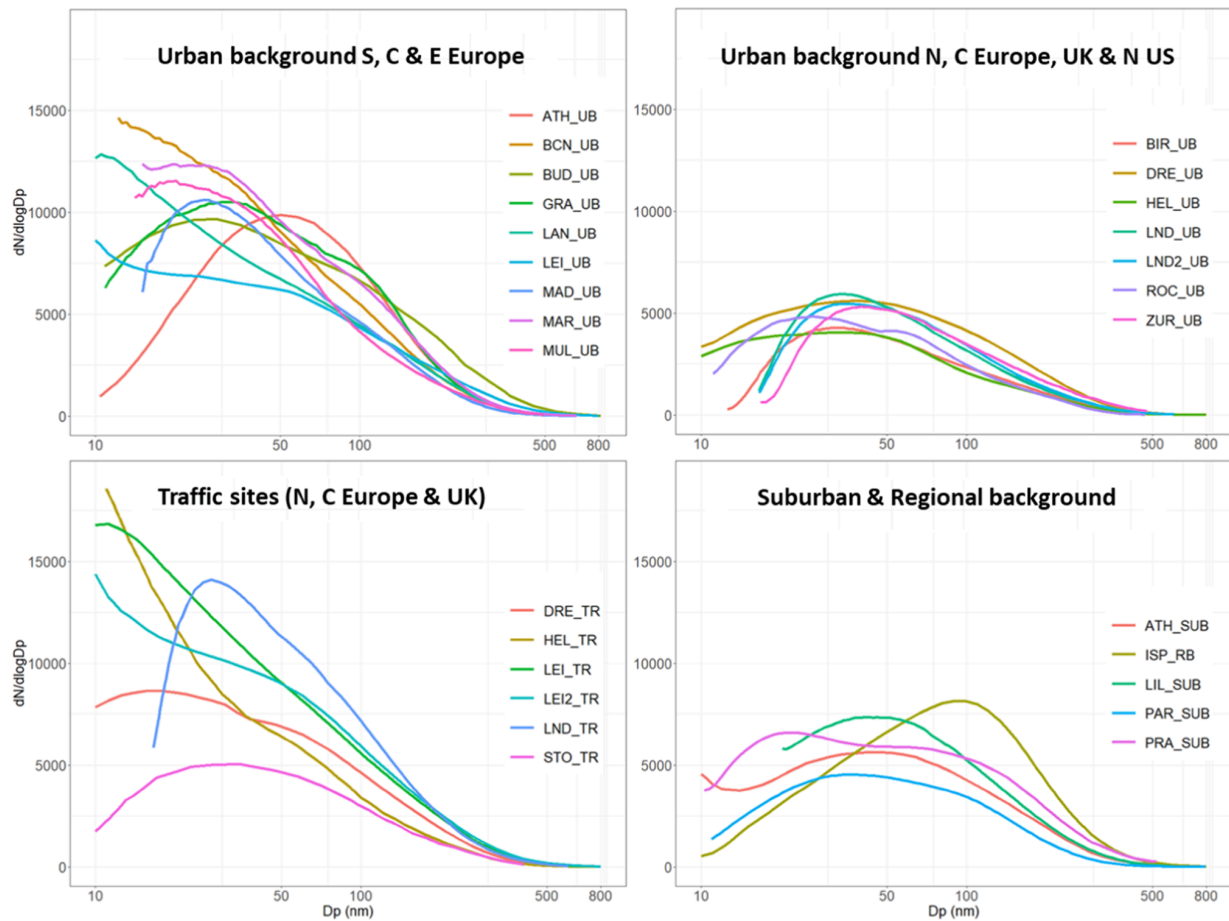


Fig. 2. Averaged 2017–2019 particle number size distributions for Traffic, Urban background sites from S, C and E Europe, Urban background from N, C Europe, UK and N US, and Suburban and Regional background sites.

Nucleation mode and a prominent size mode around 30 nm because the station is in a street canyon with high wind speeds. In this case, particles are being transported and grow due to dynamic processes (Harrison et al., 2019). However, an underestimation of the lowest size fractions cannot be ruled out due to their detection limit (17 nm).

UB sites from S, C and E Europe (ATH, BCN, BUD, GRA, LAN, LEI, MAD, MAR, MUL). Most of these sites also recorded high PNCs, but UFP-PNSDs varied widely. Thus, BCN, MAR, LEI, LAN and MUL are characterised by maxima PNC towards the finer sizes (pointing to a high impact of fresh traffic emissions and NPF). At the same time, ATH had a wide mode at 50 nm (indicating a more aged UFP and/or a relevant regional contribution (Kalkavouras et al., 2021)). The PNSDs at the remaining sites (GRA, BUD and MAD) have an intermediate prevalent mode in the aiten sizes (25–30 nm, indicating an important traffic or airport influence) despite high PNCs in the Nucleation mode.

UB sites from N and C Europe, UK and N US (BIR, DRE, HEL, LND, LND2, ROC, ZUR). These sites recorded relatively low PNCs and a prevalent Aitken mode size (30–40 nm) indicating traffic influence, or possibly airport influence as pointed out by Rivas et al. (2020), with a relatively high load of the Nucleation mode in the cases of DRE, HEL and ROC.

SUB and RB sites (ATH, LIL, PAR, PRA, ISP). These sites recorded PNCs similar to the UB sites in N Europe but with a slightly coarser prevalent mode (50 nm, indicating growth by ageing). As was expected, the PNSD of the RB site (ISP) had a prevalent 100 nm mode, due to the growth processes of the regional UFP in a highly polluted region such as the Po Valley (Sandrini et al., 2014).

3.3. Particle number concentrations

Table 3 summarises the average concentrations of UFP-PNSD, including Nucleation, Aitken and Accumulation modes (N_{10-25} , N_{25-100} and $N_{100-800}$, respectively), N_{25-800} and PNC (N_{10-800}). All concentrations are given in $\# \text{ cm}^{-3}$.

Focusing on UB sites, the highest PNCs were recorded in BCN, BUD, GRA and MAR (11,200 to 10,300 $\# \text{ cm}^{-3}$), followed by LAN, MUL, ATH, LEI, MAD and DRE (10,000 to 6200 $\# \text{ cm}^{-3}$) and LND, LND2, ROC, HEL, ZUR and BIR (4500 to 3500 $\# \text{ cm}^{-3}$) (Fig. 3). The above concentrations fall in the lower range reported by globally focused studies of UFP. De Jesus et al. (2019) reported a concentration range of 8000 to 19,500 $\# \text{ cm}^{-3}$ for 10 cities located in North America, Europe, Asia, and Australia. The concentration range reported in this present study is similar to that obtained in each of two studies based on data from four European cities (Hofman et al., 2016; Rivas et al., 2020), with 4800–11,300 $\# \text{ cm}^{-3}$. These values are very close to the $< 10,000 \# \text{ cm}^{-3}$ typical concentrations at urban background concentrations suggested by Cassee et al. (2019). Furthermore, and considering that WHO (2021b) Air Quality Guidelines states that high UFP concentrations can be considered as a health issue if daily averages exceed 10,000 $\# \text{ cm}^{-3}$, or 20,000 $\# \text{ cm}^{-3}$ on 1-h mean, many of the study sites can be considered as high UFP ones.

TR sites record high PNCs, with 13,500 to 10,500 $\# \text{ cm}^{-3}$ at the LEI, LEI2, LND and HEL sites; followed by 8800 and 5100 $\# \text{ cm}^{-3}$ at DRE and STO (Fig. 3). There are no TR sites supplying UFP-PNSD data in S and E Europe. Kumar et al. (2014) reported differences on into the cities by a factor of 2 to 9 in European cities, with higher concentrations closer to

Table 3

UFP-PNSD arithmetic averages concentrations and their standard deviations, containing Nucleation, Aitken and Accumulation mode (N_{10-25} , N_{25-100} and $N_{100-800}$, respectively), N_{25-800} and PNC (N_{10-800}) from all different stations collected in RI-URBANS. ND, Not-determined; UB, Urban Background; TR, Traffic; SUB, Suburban Background; RG, Regional Background.

Station Name	N_{10-25} # cm^{-3}	σ_{10-25} # cm^{-3}	N_{25-100} # cm^{-3}	σ_{25-100} # cm^{-3}	N_{10-100} # cm^{-3}	σ_{10-100} # cm^{-3}	$N_{100-800}$ # cm^{-3}	$\sigma_{100-800}$ # cm^{-3}	N_{25-800} # cm^{-3}	σ_{25-800} # cm^{-3}	N_{10-800} # cm^{-3}	σ_{10-800} # cm^{-3}
ATH_UB	1503	1312	5338	4642	6841	5448	1711	2060	7049	6393	8552	7064
BCN_UB	4245	4141	5560	3820	9805	6641	1380	901	6940	4441	11,186	7481
BIR_UB	661	632	2197	1813	2858	2263	673	647	2871	2318	3532	2725
BUD_UB	3691	3075	4973	3333	8664	5785	2173	1465	7146	4408	10,837	6653
DRE_UB	1935	2322	3038	2529	4973	4019	1242	1000	4280	3236	6215	4524
GRA_UB	3129	2677	5629	4160	8758	6230	1776	1431	7406	5395	10,535	7305
HEL_UB	1419	1526	2144	1764	3563	2913	644	507	2789	2056	4208	3119
LAN_UB	4628	4491	4074	2762	8702	5890	1254	888	5328	2999	9956	6657
LEI_UB	3236	4076	3524	2818	6760	5691	1403	1067	4928	3528	8163	6093
LND_UB	614	560	2983	2433	3597	2790	891	1003	3874	3223	4489	3535
LND2_UB	540	405	2912	2256	3452	2509	964	999	3875	3106	4415	3316
MAD_UB	1997	1908	4806	3632	6803	4942	1134	923	5940	4313	7937	5702
MAR_UB	2679	1760	5894	3881	8573	5160	1723	1539	7618	5161	10,297	6274
MUL_UB	2810	2139	5058	3375	7868	4955	1062	804	6120	3855	8930	5351
ROC_UB	1434	1428	2370	1849	3804	2867	602	430	2973	2105	4407	3076
ZUR_UB	296	264	2845	2156	3141	2336	1061	707	3905	2696	4201	2860
DRE_TR	3647	2939	3892	2499	7539	4655	1252	809	5144	3077	8791	5051
HEL_TR	6091	6704	3357	2816	9448	8698	1009	704	4366	3322	10,457	9088
LEI_TR	6711	5725	5272	3062	11,983	7750	1580	936	6852	3696	13,563	8134
LEI2_TR	5249	5252	5075	3368	10,324	7331	1699	1142	6774	4217	12,023	7860
LND_TR	1961	1270	6753	3943	8714	5045	1900	1261	8653	5016	10,614	6070
STO_TR	1646	1441	2756	1807	4402	2848	653	416	3409	2005	5056	3016
ATH_SUB	1654	2749	3245	2469	4899	4561	1203	725	4449	2923	6103	4892
LIL_SUB	560	625	4148	2829	4708	3230	1421	1103	5569	3557	6129	3890
PAR_SUB	1022	1055	2567	1997	3589	2789	876	708	3443	2510	4465	3241
PRA_SUB	2209	2689	3722	2636	5931	4471	1508	1101	5230	3312	7439	4875
ISP_RB	996	1231	4058	2459	5054	2924	2319	2133	6377	4189	7373	4351

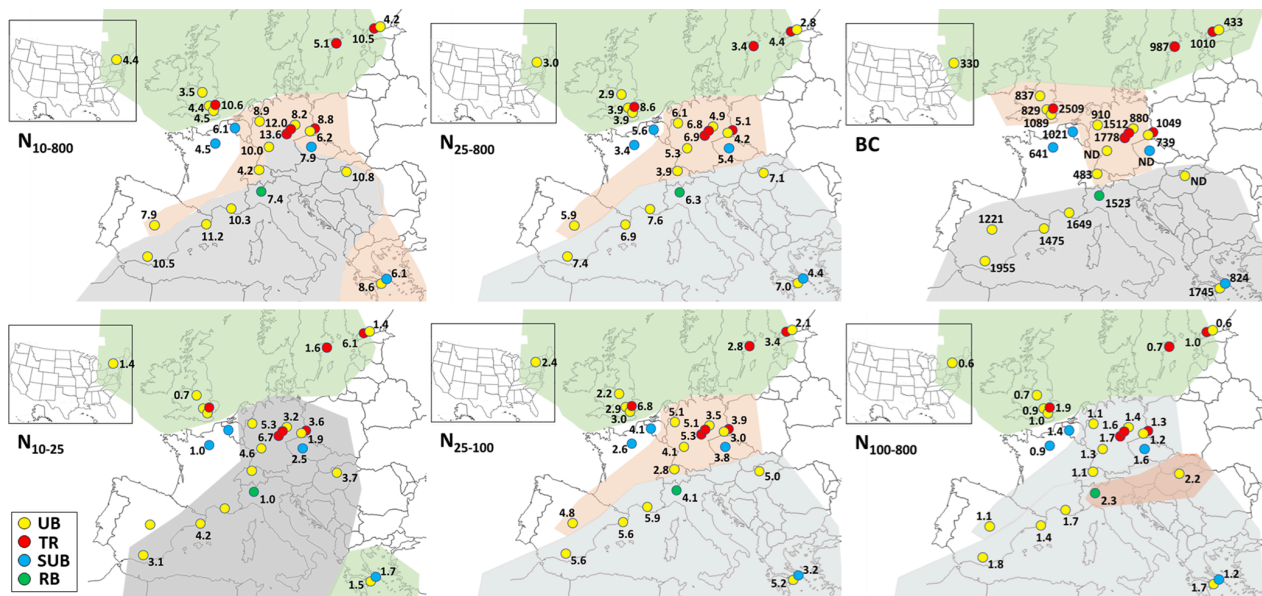


Fig. 3. Regional variability of averaged 2017–2019 particle number concentrations (in # cm^{-3} 10^{-3}) size fractions (in # cm^{-3} 10^{-3}) and black carbon (BC in ng m^{-3}) in Europe. For N_{10-25} , only sites with a lower size detection limit of 10–14 nm are included. UB, Urban background; TR, traffic; SUB, Suburban background; RB, Regional background. Shaded areas include the UB sites recording the lowest (green), intermediate (orange) and highest (grey) concentrations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the traffic sites, however in this study this factor is considerably lower (1.2 to 2.5). Moreover, [Morelli et al. \(2015\)](#), [Sánchez Jiménez et al. \(2012\)](#) and [Sun et al. \(2019\)](#) reported the high PNC concentrations for TR sites from 5 European countries, with values of 11,100 to 25,000 # cm^{-3} .

Additionally, the SUB and RB sites recorded relatively low concentrations, with 7400 to 4500 # cm^{-3} in PRA_SUB, ISP_RB, LIL_SUB,

ATH_SUB and PAR_SUB ([Fig. 3](#)). [Vratolis et al. \(2019\)](#) reported a mean value of 7600 # cm^{-3} in the size range 13 to 661 nm for the period June to August 2012 at the ATH_SUB station. The results thus show, in addition to the higher TR UFP concentrations generally reported by other studies, such as [Sánchez Jiménez et al. \(2012\)](#), [Sun et al. \(2019\)](#), [Thén and Salma \(2022\)](#), among others, a clear increasing trend of UB PNC from N (3500–4500 # cm^{-3}) to C (6200–10,000 # cm^{-3}) to S

(10,300–11,200 # cm⁻³) Europe (Fig. 3); this is probably associated with higher pollution levels, insolation (and NPF) and the impact of shipping emissions. This European spatial pattern is quite similar to that found for O₃ (EEA, 2021), although no correlation has been found between O₃ and PNCs in the present study. The increase in both PNCs and O₃ is most likely favoured by atmospheric conditions and emission patterns in S and E Europe (Carnerero et al., 2019).

When the N₂₅₋₈₀₀ values were compared to UFP concentrations (Fig. 3) without the artefact of the different lower size detection limit, a similar (to PNC) N-C-S Europe increasing trend and higher TR-UFP concentrations were again observed. Thus, TR sites reached 4400 to 8700 # cm⁻³ N₂₅₋₈₀₀, except for STO_TR (3400 # cm⁻³). For UB sites, the highest concentrations were reached in S and E European sites (5900–7600 # cm⁻³), intermediate concentrations in C European sites (3900–6200 # cm⁻³), and the lowest concentrations in N European sites, including the UK and ROC (2800–3900 # cm⁻³). The main difference found between the spatial pattern of N₂₅₋₈₀₀ and that of N₁₀₋₈₀₀ was the inclusion of ATH_UB in the S European high concentration range of N₂₅₋₈₀₀, but in the intermediate range of N₁₀₋₈₀₀, which indicates the low relevance of NPF processes in ATH compared to cities in the W Mediterranean.

Alternatively, the preservation of the N-C-S Europe increasing N-C-S Europe UFP trend when evaluating N₂₅₋₈₀₀ points to higher pollution over S and E Europe as the primary cause of this spatial trend, to a greater degree than higher NPF associated with meteorological characteristics. This finding is also supported by the spatial patterns of concentrations of BC (Fig. 3), which are very close to those of N₂₅₋₈₀₀. First, the TR sites reached the highest BC (0.99–2.7 µg m⁻³), with the highest concentrations in LND (again, the reader might consider that TR sites from S and E Europe were not included in this study). For the UB sites, the highest BC concentrations were reached again reached in S Europe (1.2 and 2.0 µg m⁻³), the intermediate concentrations (0.74–1.1 µg m⁻³) in C Europe and the UK, excluding ZUR (0.48 µg m⁻³), and the lowest concentrations (0.32–0.43 µg m⁻³) in N Europe and ROC in N US. The main difference in the spatial patterns of UB sites in Europe is the inclusion of the UK sites in the intermediate concentration range (comparable to C Europe) for BC and the low range (comparable to N Europe) for N₂₅₋₈₀₀. According to prior studies (i.e., Reche et al., 2011), the main source of BC in urban areas of European cities is road traffic (especially pre-Euro5 diesel vehicles), however the contribution of other sources such as biomass or agricultural burning cannot be excluded. The typical diurnal patterns of BC associated with hourly traffic (see below) in these cities points to road traffic as the major source.

3.4. Particle number concentrations according to size

The aforementioned differences in lower size detection limits and the correction of diffusion losses for the datasets compiled in this study was a significant drawback when comparing N₁₀₋₂₅. Thus, independently of the location and type of site, for those starting measurements at 10–14 nm the Nucleation mode reached 34 ± 11% of PNC, while for those starting at 15–20 nm it reached only 16 ± 7%. Therefore, the proportions of the Nucleation/Aitken/Accumulation modes in PNC are only discussed for those distributions starting at 10–14 nm. For this subset of sites, the highest Nucleation mode proportions in PNC were reached at the TR sites (42–58%, excluding STO, 33%). For UB sites, the lowest proportions were reached in ATH and BIR (18 and 19%, respectively), intermediate proportions (30–34%) in GRA, BUD, MUL, HEL and DRE, and the highest proportions (38–46%) in BCN, LEI and LAN. Consequently, for the UB: (i) the highest proportion of Nucleation mode concentrations was reached not only at sites from S Europe, but especially in C Europe (LEI and LAN), followed by one site in S Europe (BCN); and (ii) one S Europe site (ATH) had the lowest proportions due to the high concentration sink in this urban zone, which produced high concentrations of Aitken and Accumulation modes (Vratolis et al., 2019). In addition, moderately low Nucleation mode proportions were found at

the SUB sites of PRA, ATH and PAR (32, 27 and 23%), with the lowest at ISP_RB (14%). Accumulation mode proportions for PNC reached only 10–20% in all evaluated datasets, excluding ISP_RB at 31%. Aitken mode proportions reached 32–44% of PNC at the TR sites, excluding STO at 54%; 41–57% at the UB sites (with the highest at BIR and ATH, both 62%); and 48–57% at all SUB and RB sites.

The percentage of UFP (N₁₀₋₁₀₀) in PNC reached an average of 84 ± 3.7% for all TR, UB and SUB sites with measurements starting at sizes ranging from 10–14 nm, and 69% in the RB. The data cannot resolve the contribution of sub-10 nm aerosols. Consequently, processes such as formation of newly formed particles by nucleation occur below the detection limit (Okuljar et al. 2021; Rönkkö et al. 2017).

The higher Nucleation mode proportions for PNC in specific UB sites of C Europe (LAN and LEI), with lower insolation than in S Europe, suggest that photochemical regional or urban NPF did not exclusively drive the high proportions of the Nucleation mode, as previous work has concluded (Dall'Osto et al., 2013), but is also influenced by traffic contributions (Bousiotis et al., 2021; Harrison et al., 2011; Sun et al., 2019; Wehner et al., 2002) and the fumigation of the surface by higher-altitude atmospheric layers enriched in N₁₀₋₂₅ by NPF processes during the growth of the planetary boundary layer (PBL) (Junkermann et al., 2016); the impact of airports (Rivas et al., 2019), shipping, power plants or industrial pollution plumes might also play a significant role (Cheung et al., 2011; Diesch et al., 2013; Junkermann et al., 2016; Petzold et al., 2008; Stacey et al., 2021; Thomson et al., 2018). A relevant contribution to PNC from regional transport of UFP and the inhibition of relevant NPF might be major causes of the lowest proportion of Nucleation mode particles in ATH and coarser UFP compared with other UB sites (Kalkavouras et al., 2021).

3.4.1. Nucleation mode (N₁₀₋₂₅)

Considering only those sites with <14 nm as lower size limit for the PNSD measurements, the absolute Nucleation mode concentrations were lower at the SUB (1000–2200 # cm⁻³) and RB (1000 # cm⁻³) sites and markedly higher at the UB (1500–4600 # cm⁻³, excluding BIR_UB, with 700 # cm⁻³) and TR sites (3600–6700 # cm⁻³, excluding STO_TR, with 1600 # cm⁻³). Considering only UB sites, N₁₀₋₂₅ were higher in C, E and SW Europe (2800–4600 # cm⁻³, excluding DRE, 1900 # cm⁻³) and lower in N and SE Europe and N US (700–1500 # cm⁻³). The highest UB Nucleation mode concentrations were recorded at the following sites in the order: LAN > BCN > BUD > LEI > GRA > MUL > DRE > ATH > ROC > HEL. Therefore, as previously stated, it is not the case that S Europe (with higher insolation) records the highest concentrations of the Nucleation mode, but the contribution of other sources and processes (see above) probably significantly contributes to increasing the finest UFPs, especially in C and E Europe.

3.4.2. Aitken mode (N₂₅₋₁₀₀)

For the Aitken mode, all twenty-seven datasets were considered since all sites started measurement at lower sizes than 25 nm. Small differences were found between the Aitken mode concentrations obtained at TR sites (2800–6800 # cm⁻³) and those obtained at UB (2100–5900 # cm⁻³), SUB (2600–4100 # cm⁻³) and RB (4100 # cm⁻³) sites. However, in the last case, the reader must consider that ISP is located in the Po Valley. The lack of data from TR sites in S Europe makes it difficult to compare concentrations at TR versus UB sites across Europe. However, for UB sites, marked regional trends were again found, with the highest concentrations in S Europe (4800–5900 # cm⁻³), intermediate concentrations in C Europe (2800–5100 # cm⁻³) and the lowest concentrations in N Europe, the UK and N US (2100–3000 # cm⁻³); again, this runs parallel to the air pollution trends.

3.4.3. Accumulation mode (N₁₀₀₋₈₀₀)

The Accumulation mode concentrations at the UB sites were also higher in S Europe (1400–2200 # cm⁻³, excluding MAD, 1100 # cm⁻³) than in C (1100–1400 # cm⁻³) and N Europe, the UK and N US

(600–1000 # cm⁻³). Variability was high at the SUB and RB sites, with three SUB sites in the range 1200–1500 # cm⁻³, PAR_SUB at 900 # cm⁻³ and ISP_RB at 2300 # cm⁻³. Because the coarser detection limit varies across these datasets from 400 to 800 nm, it is difficult to make robust conclusions for this size mode.

3.5. Time variability, daily and seasonal patterns of PNC and BC

BC and PNC concentrations have parallel daily patterns, with the highest concentrations found during traffic rush hours at most sites (Fig. 4), as observed in many prior studies (Joerger and Pryor, 2018; Rivas et al., 2020; Sun et al., 2019; Von Bismarck-Osten et al., 2013;). Furthermore, a considerable number of these (LEI_UB, BCN_UB, LEI_TR and LEI2_TR, LAN_UB, MUL_UB, DRE_TR, BUD_UB, GRA_UB, ATH_UB, ATH_SUB, PRA_SUB and DRE_UB) recorded late morning-midday PNC peaks with very low BC (Fig. 4). In these cases, the intensity of the PNC midday peak varied widely, from those sites where similar concentrations to those found in the traffic rush hour peak were reached (the three LEI sites, BCN_UB, ATH_SUB, MUL_UB, the two DRE sites, BUD_UB and LAN_UB) to others where the midday peak was markedly lower than concentrations from traffic (ATH_UB, GRA_UB and PRA_SUB). Fig. 5 reveals that Nucleation mode peak concentrations, mainly caused by this midday PNC increase, were higher than those recorded during traffic rush hours for a considerable proportion of the sites (see the three LEI sites, BCN_UB, LAN_UB, MUL_UB, the two DRE sites, ATH_UB, ATH_SUB, PRA_SUB and ISP_RB). In these cases, the aforementioned photochemical nucleation and the fumigation of higher-altitude air layers rich in Nucleation mode particles, along with pollution plume impacts, made relevant contributions to PNC, and especially to N₁₀₋₂₅.

When simultaneously evaluating daily and seasonal patterns of PNC and BC, the following three types of sites were considered:

Road traffic controlling ambient PNC (N₁₀₋₈₀₀): This type of site, of which ATH_UB is an example (see bottom of Fig. 6), had a PNC peak at

traffic rush hours and a PNC which was co-variant with BC (and NO₂, NO, NO_x and CO, not shown). Furthermore, the seasonal trends of BC and PNC were very similar, with high winter and low summer concentrations. At these sites, the midday PNC peak was very weak (or absent) compared with peaks during traffic rush hours. Thus, the daily BC and PNC trends were also very similar. In this case, the highest (but still very soft) midday Nucleation mode occurred in winter-autumn, not in summer as in most cases. Furthermore, these sites were characterised by markedly lower weekend concentrations of both BC and PNC, pointing to a high impact of traffic emissions on the concentrations of both variables. The study sites with similar behaviour are GRA_UB, HEL_TR, MAD_UB, MAR_UB, PAR_SUB, ROC_UB, STO_TR and ZUR_UB (Table S3).

Conversely, some sites had **higher midday/morning PNC, than traffic rush hour**: An example is LEI_UB (see top of Fig. 6), in which the highest PNC was recorded outside traffic rush hours, at around 11–12 h. Hourly variations of averaged PNC were in parallel with O₃ (both maximising at midday) and anti-correlated with BC (PNC maximising at midday and BC being the lowest at midday and the highest at traffic rush hours). The midday PNC peak occurred (in summer, and also in spring and autumn with lower intensity) with very low BC concentrations. This can be attributed to regional or urban photo-nucleation and fumigation from higher atmospheric layers (enriched in Nucleation mode UFP and O₃ and depleted in BC, as the PBL grows by convective dynamics), and/or shipping and aviation and/or power plants and industry (see references above). In these cases, PNC maximised in summer and BC exhibited an opposite seasonal trend, with higher winter values. In most cases, weekend BC and UFP concentrations were lower. Sites included in this type are LEI_TR, LEI2_TR, DRE_UB, LAN_UB and PRA_SUB (Table S3).

Intermediate type sites (different degrees of the prevalence of traffic/midday peaks and weak seasonal patterns): An example is BCN_UB (see middle of Fig. 6). At this site, both traffic and midday peaks were present. Thus, PNC was parallel with BC except during the midday

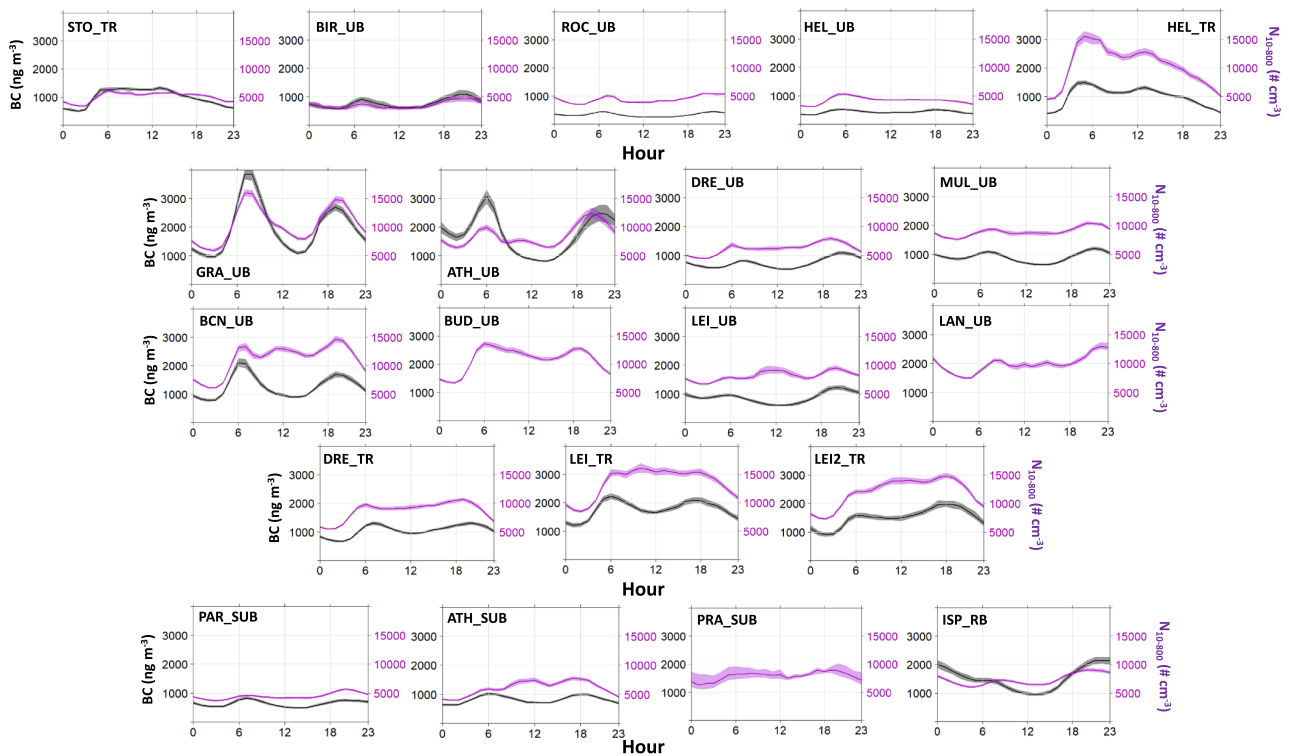


Fig. 4. Averaged 2017–2019 hourly concentrations of the particle number N₁₀₋₈₀₀ and black carbon (BC). Only sites with a lower size detection limit of 10–14 nm are included are considered. UB, Urban background; TR, Traffic; SUB, Suburban background; RB, Regional background. Particle number concentration units are # cm⁻³ 10⁻³ and those of BC in μg m⁻³.

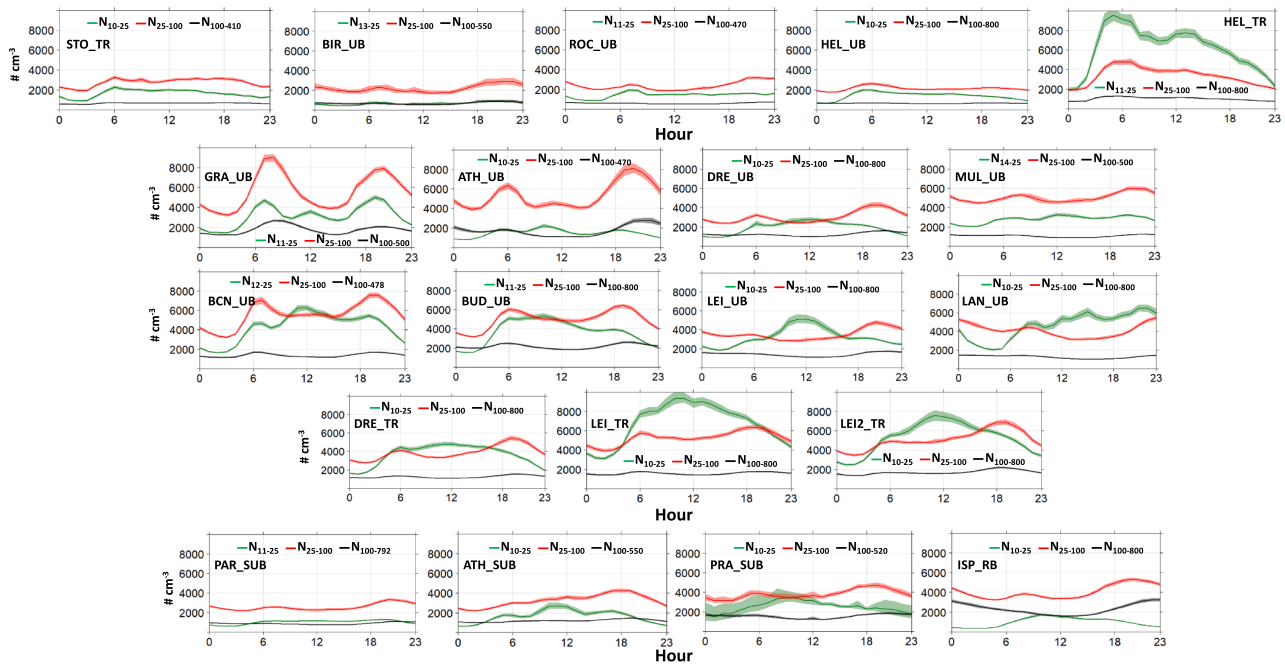


Fig. 5. Averaged 2017–2019 hourly concentrations of the particle number fractions (in $\# \text{ cm}^{-3}$) N_{10-25} , N_{25-100} and $N_{100-800}$. Only sites with a lower size detection limit of 10–14 nm are included. UB, Urban background; TR, Traffic; SUB, Suburban background; RB, Regional background.

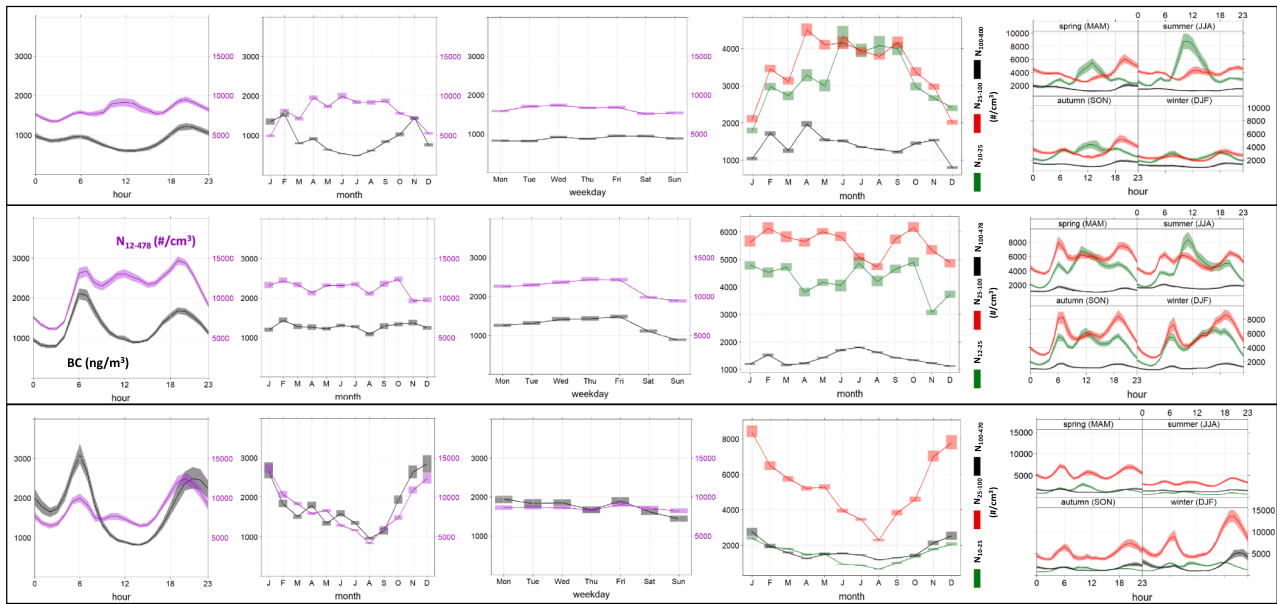


Fig. 6. Daily and seasonal patterns of UFP and BC for three Urban background stations in South Europe: LEI_UB, BCN_UB and ATH_UB.

peak, but no seasonal patterns were noticeable, with similar PNC and BC all year. These sites also had high summer Nucleation mode peaks at midday, with lower intensities in spring and autumn. Weekend BC and PNC were markedly lower. These sites had weak seasonal patterns or no covariation of BC with PNC. However, some of these sites (BIR_UB, MUL_UB, HEL_UB, LIL_SUB, LND_UB, LND2_UB and LND_TR) had greater prevalence of traffic, with weak or no midday peaks and with traffic rush hour peaks. Other stations (ATH_SUB, BUD, DRE_TR and ISP_RB) had greater prevalence of midday peaks, but did not exhibit the typical behaviour of high PNC and low BC values in summer (Table S3).

This categorisation is similar to that proposed by Sun et al. (2019) for

numerous sites in Germany.

3.6. Correlation of PNCs with ancillary pollutants and meteorological parameters

3.6.1. Black carbon

Using the twenty-four averaged dataset concentrations supplying PNC and BC data, it was observed that BC concentrations were poorly correlated with N_{10-25} ($R^2 = 0.14$, Figure S3). However, the correlation was high ($R^2 = 0.81$, Figure S3) for N_{25-100} as a result of the major traffic contribution to this mode and the absence of the effect of differences in

the lowest size detection limits used. As for BC (Fig. 4), the Aitken mode peaked at traffic rush hours (Fig. 5). This result was expected given that the reported size mode of traffic contributions to ambient urban PNC is around 40 nm. Road traffic is also the main source of BC in European cities producing very high correlations of PNC and BC during traffic rush hours (Rodríguez and Cuevas, 2007; Sun et al., 2019). However, at traffic sites, this peak could also be in the Nucleation mode (Brines et al., 2015; Harrison et al., 2019, 2011; Hopke et al., 2022; Rivas et al., 2020; Wehner et al., 2002). The correlation of BC with $N_{100-800}$ was also high but lower than for N_{25-100} ($R^2 = 0.65$, Figure S3), indicating a major traffic origin with partial contributions of other sources, such as regional aerosol transport (among others).

Correlation of BC with PNC (N_{10-800}) and N_{25-800} was high, with $R^2 = 0.60$ and 0.85 , respectively, when including the averages of all sites; $R^2 = 0.26$ and 0.89 for TR sites; $R^2 = 0.71$ and 0.85 for UB sites; and $R^2 = 0.85$ and 0.90 for SUB and RB sites (Figure S4). High correlations between these factors were also found by Sun et al. (2019) for numerous sites in Germany. The high correlation of average BC with N_{25-800} reflects the impact of traffic pollution on air quality; thus, cities which are highly polluted have higher concentrations of co-pollutants than cleaner cities. However, when considering N_{10-800} and N_{10-25} (Figures S3 and S4), the correlation decreased markedly because (i) the Nucleation mode particles have mixed origins (traffic, nucleation, aviation, shipping, fumigation for upper atmospheric layers, and biomass burning, among others) and (ii) the differences in the lower size detection limits are considerable. When correlating BC and PNC individually for each site using hourly data for 2017–2019, high correlation ($R^2 = 0.60–0.66$) was obtained for three sites, and moderate correlation ($R^2 = 0.39–0.57$) for the other ten sites; while for the other eleven sites, the correlation was low ($R^2 = 0.18$ with an exceptionally small value at LEI UB of $R^2 = 0.01$) (Table S4). When considering N_{25-800} instead of PNC, the correlation with BC markedly increased in such a way that the correlation was high for six sites ($R^2 = 0.60–0.78$), moderate for another ten sites ($R^2 = 0.40–0.46$) and lower in the remaining eight sites ($R^2 = 0.26–0.34$, with the exception of LEI UB at $R^2 = 0.05$) (Table S4).

The correlation of N_{25-100} and $N_{100-800}$ with BC was high ($R^2 = 0.72$ and $0.62–0.88$) for one and nine sites, respectively; and moderate ($R^2 = 0.40–0.56$ and $0.40–0.58$) in the other eight and thirteen sites, respectively, of the twenty-four sites that measure UFP and BC (Table S5). For N_{10-25} , the correlation was very low in nineteen out of the twenty-four cases ($R^2 = 0.04–0.10$) and low to moderately low in the other five ($R^2 = 0.16–0.36$) (Table S5).

The above correlation ranges are again similar to those found by Sun et al. (2019) and Wu et al. (2015) in Germany and streets in Edinburgh during different campaigns and at different sites, and it is essential to highlight that:

- The correlation of BC with the mean N_{10-800} and N_{25-800} values of the sites indicates that these equally reflect the impact of road traffic emissions on air quality;
- The correlation analysis of individual sites shows that for BC vs. N_{10-800} 69% of the sites reached moderately-high (39%) and moderately-low (35%) correlations, but for 26% of the sites the correlation was low;
- For N_{25-800} the correlation with BC increased at 96% of the sites (high 70%, moderate 26%);
- Adequate monitoring of the Nucleation mode UFPs might supply information very relevant to monitoring the impact of other sources and processes.

However, it is recommended that future studies:

- Start measurements at sizes even lower than 10 nm by using instruments such as co-located Particle Size Magnifiers (PSM) or Nano-CPCs;
- Meet ACTRIS and CEN requirements on size measurements (N_{10-800}).

This will allow harmonised and systematic data compilation for, e.g., aerosol health studies, more detailed process-level studies, and better interpretation of the results.

3.6.2. Other ancillary pollutants and meteorological parameters

As for the other ancillary pollutants, low correlations were found between PNC and CO and SO_2 , when considering the 2017–2019 individual averages (Table S6), only for UB sites ($R^2 = 0.21$ for both CO and SO_2). When considering N_{25-800} rather than PNC, the correlation was the same for CO and slightly higher for SO_2 ($R^2 = 0.21$ and 0.42 , respectively); and for CO, a moderately-low correlation ($R^2 = 0.36$) was also found for TR sites. For the individual sites (Table S4), low to moderate correlations with PNC ($R^2 = 0.17–0.54$) were found only for five sites out of the seventeen co-measuring SO_2 ; but low to moderately-high correlations with PNC ($R^2 = 0.16–0.65$) were found in eleven of the sixteen sites co-measuring CO. For N_{25-800} , the correlation did not increase for the same five sites with low to moderate correlation ($R^2 = 0.13–0.53$) for SO_2 and the same eleven sites with low to moderate-high correlation ($R^2 = 0.13–0.71$) for CO.

For NO and NO_2 , moderate-low correlations were obtained with PNC when considering the averaged concentrations (Table S6) of only UB sites ($R^2 = 0.27$ and 0.35 , respectively); but these increased for N_{25-800} at TR ($R^2 = 0.70$ and 0.63 for NO and NO_2 , respectively) and UB ($R^2 = 0.35$ and 0.48) sites. For SUB and RB sites, significant moderate-low to moderate-high correlations were attained for PNC and N_{25-800} with NO ($R^2 = 0.23$ and 0.63). This is because urban NO_x and BC are emitted at a high proportion by diesel vehicles (Degraeuwe et al., 2016; Ježek et al., 2015; Tunno et al., 2018). Furthermore, Harrison and Jones (2005) reported similar correlations between PNC and NO_x at one TR site (Marylebone Rd., with $R^2 = 0.63$) and at several UB sites (Belfast Centre, Glasgow Centre, Birmingham Centre and London Bloomsbury, with $R^2 = 0.37$, 0.39 , 0.46 and 0.43 , respectively). A higher correlation of both NO and NO_2 with N_{25-800} was thus expected at TR sites.

When considering the individual datasets (Table S4), low-moderate correlations between PNC with NO_2 and NO ($R^2 = 0.22$ to 0.58 , slightly higher for NO_2) were obtained only at TR sites. For the sixteen UB sites co-measuring NO_2 and NO, only twelve and nine, respectively, reached moderate-low to moderate-high correlations ($R^2 = 0.23$ to 0.68 for NO_2 and $R^2 = 0.21$ to 0.54 for NO), while for SUB and RB sites, correlations decreased and varied greatly ($R^2 = 0.07$ to 0.40) for both pollutants. Similar results were obtained for N_{25-800} with NO_2 and NO, with slightly lower correlations of NO and NO_2 at TR sites ($R^2 = 0.11$ to 0.56), but higher correlations at UB sites: all UB sites had $R^2 = 0.16–0.69$ for NO_2 and twelve out of fourteen UB sites had $R^2 = 0.16$ to 0.56 with NO. Again, for the SUB and RB sites, correlations varied greatly ($R^2 = 0.06$ to 0.44) for both pollutants. Thus, as for BC, the results show that in most UB and TR sites, there was a significant co-hourly variation of NO_2 and NO with PNC and N_{25-800} .

For PNC and PM_{10} (Table S6), the correlation was negative and moderate-low for TR sites ($R^2 = 0.32$) and was positive and moderate-low to moderate-high for UB and SUB + RB sites ($R^2 = 0.37$ and 0.68). For N_{25-800} , the correlation markedly increased for UB and SUB + RB sites ($R^2 = 0.53$ and 0.91) but not for TR sites ($R^2 = 0.00$). For $PM_{2.5}$ and PNC (Table S6), the correlation increased from TR ($R^2 = 0.05$) to UB ($R^2 = 0.40$) and to SUB + RB ($R^2 = 0.77$); and increased when considering N_{25-800} ($R^2 = 0.77$ TR, 0.49 UB and 0.91 SUB + RB). Thus, when the regions/sites of Europe recorded higher $PM_{2.5}$ concentrations, PNC, especially of N_{25-800} , was also high.

However, the results of evaluations of individual sites (Table S4) differ considerably, -with low PNC- PM_{10} correlations: only eight sites reached $R^2 = 0.16–0.36$ and sixteen reached $R^2 = 0.00–0.12$. For N_{25-800} , the correlation with PM_{10} increased slightly, with twelve sites reaching $R^2 = 0.16–0.45$ and twelve reaching $R^2 = 0.00–0.14$. A similar situation was found for $PM_{2.5}$ and PNC correlations, with only four sites reaching $R^2 = 0.19–0.34$ and thirteen reaching $R^2 = 0.00–0.14$. Again, for N_{25-800} the correlation increased slightly, with seven sites reaching

$R^2 = 0.17$ – 0.47 and ten reaching $R^2 = 0.04$ – 0.15 . For the different modes, as expected, the correlations of the Nucleation mode concentrations with PM_{10} and $PM_{2.5}$ were significantly lower at all sites ($R^2 = 0.00$ – 0.11 and 0.00 – 0.07 , respectively, Table S5). For the Aitken mode, low-moderate correlations were only reached at four sites for PM_{10} ($R^2 = 0.20$ – 0.22) and $PM_{2.5}$ ($R^2 = 0.16$ – 0.31) and for all other sites, correlations even decreased: $R^2 = 0.00$ – 0.14 and 0.01 – 0.13 , respectively (Table S5). The correlation increased for the Accumulation mode with PM_{10} , at $R^2 = 0.16$ – 0.64 ($R^2 = 0.04$ – 0.15 for the remaining five sites). All seventeen sites measured correlations with $PM_{2.5}$ reaching $R^2 = 0.19$ – 0.79 (Table S5). De Jesus et al. (2019) found low correlations of $PM_{2.5}$ and PNC in a global study involving ten cities located in North America, Europe, Asia, and Australia, with r^2 = from 0.01 to 0.48.

Using the averaged concentrations for the different sites, correlations were low for O_3 with PNC and N_{25-800} (Table S6) for UB and SUB + RB sites ($R^2 < 0.07$), with the exception of N_{25-800} at SUB + RB sites ($R^2 = 0.24$). Markedly higher negative PNC and N_{25-800} O_3 correlations were obtained for TR sites, at $R^2 = 0.72$ and 0.91 , respectively. However, for the individual sites (Table S4), the correlation of PNC and N_{25-800} with O_3 was low and negative in all cases, with only seven and eight out of twenty-three sites reaching $R^2 = 0.19$ to 0.39 , respectively, and much lower correlations in all the other cases.

When using 2017–2019 average data for meteorological parameters from all sites, a moderate-low to moderate positive correlation was obtained for both PNC and N_{25-800} with T and insolation ($R^2 = 0.28$ to 0.53) and a negative correlation with relative humidity ($R^2 = 0.27$ to 0.46) was obtained for UB sites (Table S6). This result is probably a product of the gradient from the low UFP pollution in N Europe to higher UFP pollution in S and E Europe. When using the single site hourly datasets (Table S4), correlations were found to be low for all parameters evaluated (mostly from negative to positive, $R^2 = 0.09$), with a more positive trend for Nucleation mode vs. insolation and a more negative trend for wind speed with all PNC size modes, relative humidity, and Nucleation mode particles (Table S5).

4. Conclusions and limitations

This work summarises the first results on ultrafine particle (UFP) particle-size distribution (PNSD) in urban areas of Europe from the RI-URBANS project. This includes 2017–2019 datasets from twenty-seven air quality monitoring sites. The focus is mostly on urban background (UB) and traffic (TR) concentrations, but it also includes four suburban (SUB) and a regional background (RB) sites.

4.1. Measurements

Only fourteen out of twenty-seven datasets reached >70% of data availability, reflecting the complexity of UFP-PNSD measurements, which require detailed monitoring and constant maintenance of the instrumentation. Furthermore, the lack of harmonisation of PNSD measurement accounts for significant differences of the lower size detection limits (3 to 20 nm), which makes the direct comparison of UFP and particle number concentrations (PNC) concentrations from different sites difficult, especially those in the Nucleation mode (<25 nm). To increase comparability, an effort should be made to implement the ACTRIS and CEN recommendations in the near future.

From the twenty-seven datasets, only twelve are openly available in the EBAS data infrastructure, and accordingly, an effort should also be directed at making data open for use in air quality health research and policy support, especially if the data is obtained with public funds.

4.2. Levels and PNSD

The results show a clear increasing N_{10-800} UB concentration trend from Northern to Southern and Eastern Europe, and an even more marked trend for N_{25-100} and N_{25-800} . These trends are similar to those

found for Black Carbon (BC). As it could be expected, PNCs follow the TR > UB > SUB trend. However, PNC in the RB of the Po Valley are equivalent to those of an UB in Southern Europe.

The major contributing source to PNC in urban Europe is road traffic as deduced from the parallel daily patterns to those of BC, with peak concentrations at traffic rush hours. However, a relevant number of sites recorded high PNCs (in some cases exceeding those of traffic) in the morning-midday coinciding with minimal BC, and the highest temperature, wind speed, insolation and O_3 . These marked morning-midday maxima are caused by pronounced increases of N_{10-25} (Nucleation mode).

It is important to note that the highest midday PNC-Nucleation modes are recorded in Central Europe followed by Southern Europe, so they do not follow only insolation as it should be expected from new particle formation from regional photochemical nucleation. Thus, the origin of these midday marked maxima are attributed to: photochemical nucleation, surface fumigation of high-altitude atmospheric layers transporting high SO_2 plumes and plumes from airports, shipping, or power or industrial plants. In a relevant proportion of the study sites, the contribution of these sources/processes to the annual PNC is very significant.

Daily and seasonal patterns are used to classify the PNSD datasets in three major groups: i) low midday PNC peaks and PNC parallel daily patterns, with high winter BC and PNC, with low spring-summer N_{10-25} ; ii) PNC and BC traffic-related daily patterns, but a major midday PNC peak with low BC concentrations, with inverse seasonal PNC (highest in spring-summer) and BC (highest in autumn-winter) patterns, with very intensive N_{10-25} midday peaks in spring and summer; and iii) as type ii, but with dominant traffic-related PNC peaks, but marked midday N_{10-25} peaks and no definite seasonal patterns.

Strong correlations between averaged BC and PNC concentrations, especially for BC and N_{25-800} , of the different datasets indicate that there is a clear impact of road traffic emissions on air quality in UB environments, with different degree across Europe. However, for the individual datasets BC-PNC are notably lower, because i) there are more sources contributing to PNC than traffic; ii) the high midday PNC in many of the sites decrease BC-PNC correlation. Moderate-correlations are also attained for the individual sites for N_{25-800} and NO and NO_2 for UB and TR sites.

These findings support the need of measuring PNCs accurately, especially the Nucleation mode, to properly evaluate the occurrence of sources other than traffic contributing to the health effects of UFP. To this end, RI-URBANS strongly recommends starting measurements below 10 nm by using instruments such as co-located Particle Size Magnifiers (PSM) or Nano-CPCs and implementing the above recommended protocols for UFP-PNSD measurements.

Further steps in the study of these datasets will focus on source apportionment analyses and the epidemiological assessments to evaluate short-term associations with PNCs, size modes and source contributions.

4.3. Limitations

As a major limitation we should acknowledge the problems in directly comparing the datasets due to different protocols used for PNSD measurements and quality assurance. Thus, the DRE_UB, LAN_UB, LEI_UB, DRE_TR, LEI and LEI2_TR datasets were obtained following the ACTRIS protocols for measurements of PNSD, and the instruments are frequently undergoing quality assurance exercises by the World Calibration Centre for Aerosol Physics (WCCAP, <https://www.eurochamp.org/calibration-centres/wccap>). The datasets from ATH_UB, BCN_UB, GRA_UB, HEL_UB, MAD_UB, HEL_TR, ATH_SUB, LIL_SUB, PRA_SUB, ISP_RB are produced by ACTRIS collaborators or partners, and they are obtained with protocols tending to follow the ones from ACTRIS but have not recently or have never undergone the quality checks by the WCCAP. Finally, the datasets of BIR_UB, BUD_UB, LND and LND2_UB,

MAR_UB, ROC_UB, ZUR_UB, PAR_SUB, LND_TR and STO_TR are obtained from highly specialized research teams, but data uncertainty remains unknown. The lack of harmonisation has caused variability in the lower size detection limit leading to large differences in the total number of measured particles. In this study, this discrepancy is reduced by comparing concentrations in the range of 10–800 and 25–800 nm, but there is an urgent need to harmonise measurement protocols such as following ACTRIS and CEN recommendations to provide directly comparable UFP-PNSD datasets.

Credit authorship contribution statement

The corresponding author, Pedro Trechera, is responsible for ensuring that the descriptions are accurate and agreed upon by all authors, and was involved in data collection, treatment and processing. Also, is the responsible to write the manuscript.

Meritxell Garcia-Marlès was involved into data treatment, data processing and writing, including the revision of the manuscript.

Xavier Querol is the coordinator of the project and accordingly, he did conceptualisation, supervision, but also as member of the writing lead team of this paper was involved data treatment, data processing and writing, including the revision of the manuscript.

Xiansheng Liu did the correlation of UFP-PNSD for each individual dataset from 2017–2019 with BC, ancillary pollutants and meteorological parameters.

Roy M. Harrison, Philip K. Hopke, Tuukka Petäjä and Andrés Alastuey provided the 2017–2019 datasets for the different sites, assisted and contributed in the processing of the dataset. In addition, they carried out a thorough proofreading of the manuscript before obtaining the final version of the manuscript.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share the data from all the sites. However, should there be a request for the data, every effort will be made to share it.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2023.107744>.

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4.2.2 Paper II

Inter-annual trends of ultrafine particles in urban Europe

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Inter-annual trends of ultrafine particles in urban Europe

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ABSTRACT

Ultrafine particles (UFP, those with diameters ≤ 100 nm), have been reported to potentially penetrate deeply into the respiratory system, translocate through the alveoli, and affect various organs, potentially correlating with increased mortality. The aim of this study is to assess long-term trends (5–11 years) in mostly urban UFP concentrations based on measurements of particle number size distributions (PNSD). Additionally, concentrations of other pollutants and meteorological variables were evaluated to support the interpretations. PNSD datasets from 12 urban background (UB), 5 traffic (TR), 3 suburban background (SUB) and 1 regional background (RB) sites in 15 European cities and 1 in the USA were evaluated. The non-parametric Theil-Sen's method was used to detect monotonic trends. Meta-analyses were carried out to assess the overall trends and those for different environments. The results showed significant decreases in NO, NO₂, BC, CO, and particle concentrations in the Aitken (25–100 nm) and the Accumulation (100–800 nm) modes, suggesting a positive impact of the implementation of EURO 5/V and 6/VI vehicle standards on European air quality. The growing use of Diesel Particle Filters (DPFs) might also have clearly reduced exhaust emissions of BC, PM, and the Aitken and Accumulation mode particles. However, as reported by prior studies, there remains an issue of poor control of Nucleation mode particles (smaller than 25 nm), which are not fully reduced with current DPFs, without emission controls for semi-volatile organic compounds, and might have different origins than road traffic. Thus, contrasting trends for Nucleation mode particles were obtained across the cities studied. This mode also affected the UFP and total PNC trends because of the high proportion of Nucleation mode particles in both concentration ranges. It was also found that the urban temperature increasing trends might have also influenced those of PNC, Nucleation and Aitken modes.

1. Introduction

Atmospheric particulate matter (PM) is one of the main air pollutants (WHO, 2021a). It is estimated that ambient particulate air pollution caused 4.2 million premature deaths worldwide in 2019, as derived from the aggravation of cardiovascular and respiratory diseases, among others (WHO, 2021a). The nature and severity of the effects depend, among others, on the size of the particles. Therefore, air quality standards are set up for the ambient air mass concentrations of particles ($\mu\text{g m}^{-3}$) with aerodynamic diameters $\leq 10 \mu\text{m}$ (PM₁₀) and $\leq 2.5 \mu\text{m}$ (PM_{2.5}). However, a large fraction of the particles in the atmosphere are smaller than 100 nm (diameters can be as fine as 1 nm). These very fine particles contribute minimally to the PM mass concentrations but represent the bulk of the particle number concentrations (PNC, # cm⁻³), which are not regulated by ambient air quality standards (Seinfeld and Pandis, 2016). Thus, when focusing on this aerosol size range, measurements are performed as PNC and not PM mass. PNC measurements are frequently derived from particle number size distribution (PNSD) by calculating PNCs for specific size ranges. Conventionally, the term ultrafine particles (UFP) is used for particles with a diameter ≤ 100 nm, while the Nucleation, Aitken and Accumulation size modes refer to particles in the <25, 25–100 and >100 nm size ranges, respectively (e.g., Seinfeld and Pandis, 2016; CEN, 2020; ISO, 2023). The term “nanoparticle” is usually used for manufactured particles, while “ultrafine” is outdoor or indoor aerosols of a similar size. Furthermore, because most of the total PNC is accounted for by UFPs in urban areas (84 ± 3.7 % as average and standard deviation of 27 datasets in Europe, Trechera et al., 2023), the terms PNC and UFP are used in many studies as equivalents. Thus, PNC and UFP are not actually equivalents, but PNC is used as a surrogate for true UFP.

It has been reported that the size of UFPs not only allows them to reach the deeper parts of the respiratory system but also to translocate through the alveoli and reach the circulatory system and from there any organ in the body (Oberdörster et al., 2005; Peters et al., 2006; Kreyling et al., 2014; Cassee et al., 2019). Furthermore, other studies also evidenced that there is translocation through the olfactory nerve in the nose into the brain, and UFP have been reported to cross cell membranes in ways other than phagocytosis (Oberdörster et al., 2004; Genc et al., 2012; among others). A number of studies found a positive relationship between exposure to UFP and increased premature mortality (e.g., Wichmann et al., 2000; Ibaldo-Mulli et al., 2004; Tobías et al., 2018; Schwarz et al., 2023). However, Ohlwein et al. (2019), US EPA (2019) and WHO (2021b) concluded that the epidemiological results of the

health effects of UFP are inconsistent. This inconsistency might be due to different types of UFP prevailing in different regions, a lack of harmonization in UFP measurements, high exposure assignment error based on the high microscale variability in space and time of UFP concentrations, a lack of long-term time-series available to epidemiological studies, or a lack of actual widespread impacts on health (Cassee et al., 2019). Based on the above considerations, the last WHO Air Quality Guidelines did not set up specific guidelines for PNC (or UFP), but recommended the long-term measurements of these variables with harmonized measurement protocols to better evaluate the convenience of such specific guidelines for UFP (WHO, 2021b). Alternatively, CEN (2020), ACTRIS (2021), RI-URBANS (2022) and Trechera et al. (2023) suggest the measurement of total PNCs with at least a finer size detection limit of 10 nm (50 % efficiency) and PNSD measurements in the range of 10 to 800 nm. PNC in the range of <10 nm might be high, but measuring this fraction is complex, and it is also recommended by ACTRIS (2021) and RI-URBANS (2022) in areas where nucleation mode particles are expected to be high.

Shipping is the major source of PNC in Europe with around $8.2 \cdot 10^{26}$ # yr⁻¹ followed by road traffic with $4.3 \cdot 10^{26}$ # yr⁻¹, and industry with $4.2 \cdot 10^{26}$ # yr⁻¹ (Kuenen et al., 2022). However, as occurs also for NO₂, the proximity of the urban population to road traffic emission accounts for a very high road traffic contribution to urban PNC exposures. Thus, studies on source apportionment of UFP point to road traffic as the main source in urban areas (Zhou et al., 2005; Pey et al., 2009; Dall'Osto et al., 2011; Harrison et al., 2011; Brines et al., 2014, 2015; Beddows et al., 2015, 2019; Rivas et al., 2020; Hopke et al., 2022). Traffic typically accounts for >70 % of the annual UFP emissions in urban environments (Hopke et al., 2022). In addition to road traffic, photochemical nucleation (leading to new particle formation), urban background (with a high load from traffic), regional background, long-range transport, harbours, airports, and domestic heating emissions have been reported to contribute to urban UFP concentrations. As it could be expected from its origin, photochemical nucleation is characterised by PNSD peaking at the finest sizes, followed by harbours and airports, typically with a prevailing Nucleation mode, see for example Rivas et al. (2020) and Stacey et al. (2020). Traffic exhaust emissions typically result in two major modes at 30–35 nm and 60–80 nm (Hopke et al., 2022), but also a major fraction of very small particles (smaller than 30 nm in diameter) (Trechera et al., 2023). The remaining aforementioned sources (urban and regional background, long range transport and domestic heating) are characterised by modes close to or coarser than 100 nm, according to the above references.

The few available studies analysing long term trends of UFP (e.g., Simon et al., 2020; Mikkonen et al., 2020; Presto et al., 2021; Chen et al., 2022; Damayanti et al., 2023) have found decreasing trends over the last two decades. Given that road traffic is the major source of urban UFP, these trends have been related to the reduction of particulate emissions from vehicle exhausts by the implementation of emission standards, especially EURO 5 and 6 (for cars) and V and VI (for heavy duty vehicles), and the equivalent TIER standards (2 and 3) in US, for which diesel particle filters (DPF) are required. The decrease of the sulfur content in the fuels of the vehicles might have also had an important impact in abating SO₂, and associated nucleation of particles (Chen et al., 2022), but this was already implemented in Europe before the period of this study. Damayanti et al. (2023) found that for a street canyon in London this effect was very evident for the Aitken and Accumulation modes, but the Nucleation one was largely unaffected. They proposed that the DPF do not retain semi-volatile organic compounds (SVOCs) emitted by diesel cars, and the nucleation of these SVOCs and the abatement of the condensation sink (CS) might account for the non-decreasing trend of the Nucleation mode. Furthermore, abrasion UFP are also emitted as part of the non-exhaust vehicle emissions (Dahl et al., 2006; Mathissen et al., 2011; Niemann et al., 2020; Li et al., 2023; and references therein).

Based again on the European emission inventory for the PNC-PNSD, 2018 emissions from diesel passenger cars reached $17.5 \cdot 10^{25} \text{ \# yr}^{-1}$, while emissions from petrol cars reached around $0.5 \cdot 10^{25} \text{ \# yr}^{-1}$ (Kuenen et al., 2022). Furthermore, emissions of $9.8 \cdot 10^{25} \text{ \# yr}^{-1}$, and $13.5 \cdot 10^{25} \text{ \# yr}^{-1}$ from (mostly diesel) light and heavy-duty vehicles have to be added. Thus, the diesel fleet is still responsible for a very large fraction of UFP emissions in Europe.

The implementation of major air quality measures and the EURO/TIER standards have also had an effect on the abatement of black carbon (BC) (Savadkoobi et al., 2023), and nitrogen dioxide (NO₂) and carbon monoxide (CO) (EMEP, 2016; ETC, 2020). Concentrations of SO₂ (mainly emitted by fossil fuel burning in Europe) drastically decreased as a consequence of deindustrialization and regulation measures implemented at European scale (EMEP, 2016). On the other hand, the concentrations of PM₁₀ and PM_{2.5} also decreased in 2000–2010 but a flat trend was recorded since then at many sites (EMEP, 2016; ETC, 2020). In contrast, concentrations of O₃ tend to increase in many urban environments due to the reduction of nitrogen oxides (NO_x) in a VOC-limited urban O₃ formation regime or to a steeper decrease of nitrogen monoxide (NO) versus NO₂ and the consequent lower titration of O₃ in environments without strong local O₃ formation (ETC, 2020; Sicard et al., 2021).

RI-URBANS (Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial Areas, the European Union's Horizon 2020 research and innovation program, contract 101036245) is a European research project, which demonstrates the applications of advanced air quality service tools in urban Europe to improve the assessment of air quality policies, including a better evaluation of health impacts. In this context, this study aims to gather and evaluate available long-term trends of UFP and different particle size modes, based on PNSD data obtained at 21 (mostly urban) sites from 15 urban areas in Europe (11 member states) and one from the USA. A prior RI-URBANS study (Trechera et al., 2023) reported the phenomenology of UFP in urban Europe and compared the 2017–2019 concentrations across different cities and environments. Additional work is ongoing on the source apportionment of PNSD for 23 (mostly urban) sites using receptor modelling tools. In the current study we evaluate and interpret the trend analyses of the above 21 datasets for a period covering up to 11 years (2009–2019). A number of sites cover a shorter period of time, but in all cases, at least the last 5 years of measurement are available. The years 2020–2022 are intentionally excluded to avoid interferences of the COVID19-lockdown effects. To support interpretations, we also evaluate compiled datasets of conventional pollutants (BC, NO₂, NO, CO, SO₂, O₃, PM₁₀, PM_{2.5} and PM₁) and

meteorological variables (temperature, T; relative humidity, RH; atmospheric pressure, P; wind speed, WS; solar radiation, RAD; and rain, R) concurrently measured with the PNSDs.

2. Methodology

2.1. Cities and sites providing UFP-PNSD data

This study is based on 5 to 11 years UFP-PNSD datasets from the period 2009–2019 compiled by RI-URBANS and supplied from advanced air quality monitoring networks and research sites. The selection of sites for this study is based upon a previous study on the phenomenology of UFP in urban Europe by Trechera et al. (2023), allowing coverage of all ranges of concentrations of urban pollutants in cities from regions with different climate patterns, emission volume and sources and urban morphologies. These include (see Fig. 1 and Table 1):

- Twelve urban background (UB) sites covering most of Europe: Athens (ATH_UB), Barcelona (BCN_UB), Budapest (BUD_UB), Dresden (DRE_UB), Helsinki (HEL_UB), Lecce (LEC_UB), Leipzig (LEI_UB), London (LND_UB), Madrid (MAD_UB), Mülheim (MUL_UB), Zurich (ZUR_UB), and Rochester (ROC_UB) in New York State in US.
- Five traffic (TR) sites in Central (C), West (W) and North (N) Europe: Dresden (DRE_TR), Helsinki (HEL_TR), Leipzig (LEI_TR), London (LND_TR) and Stockholm (STO_TR); but not TR sites in South (S) Europe.
- Three suburban background (SUB) sites and one regional background (RB): Athens (ATH_SUB), Paris (PAR_SUB) and Prague (PRA_SUB); the regional background of Ispra (IPR_RB).

Thus, in total, 21 sites provided at least 5 years of hourly data in the period 2009–2019 of UFP-PNSD for this study. The period was selected excluding 2020–2022 to avoid the effects of the decrease of pollution due to the COVID19-lockdown, which were variable across urban Europe (Salma et al., 2020; Dinoi et al., 2021; Eleftheriadis et al., 2021; Petit et al., 2021; Putaud et al., 2021 and 2023). All the PNSD data used in this study is openly available in the EBAS database infrastructure (<https://ebas.nilu.no/>). Data corresponding to co-located measurements of other pollutants (BC, NO₂, NO, CO, SO₂, O₃, PM₁₀, PM_{2.5} and PM₁) and meteorological variables (T, RH, P, WS, RAD and R) were also collected, when available, from the same sites or nearby air quality monitoring sites. Table S1 summarises the hours/year of UFP-PNSD measurements included in this study for each site.

2.2. Instrumentation and measurements

PNSD datasets were obtained from different types and models of Mobility Particle Size Spectrometer (MPSS), reported in Table 2. Concentrations of BC, PM and gaseous pollutants, and meteorological variables were collected from each site by instrumentation fulfilling European standards. All the data were averaged to hourly values.

Since different instruments were used to obtain PNSD datasets, the measured size ranges varied. Following ACTRIS and CEN guidance, it is suggested to measure the total PNCs with at least a finer size detection limit of 10 nm, and the recommended size measurement range for PNSD is 10–800 nm. As reported in Trechera et al. (2023), several sites start size measurements from 11 to 17 nm (BCN_UB, BUD_UB, HEL_TR, LND_UB, LND_TR, MAD_UB, MUL_UB, PAR_SUB, ROC_UB and ZUR_UB), instead of the recommended 10 nm, or end with lower coarser sizes from 410 to 794 nm (ATH_UB, ATH_SUB, BCN_UB, LND_UB, LND_TR, MAD_UB, MUL_UB, PAR_SUB, PRA_SUB, ROC_UB, STO_TR, and ZUR_UB), instead of the recommended 800 nm. The latter does not have a big impact to the total PNC because of the relatively low PNC within the coarser sizes. However, the lower size detection limit affects to a much higher degree the total PNC, and especially the Nucleation mode concentrations. Thus, when comparing PNSD concentrations from all

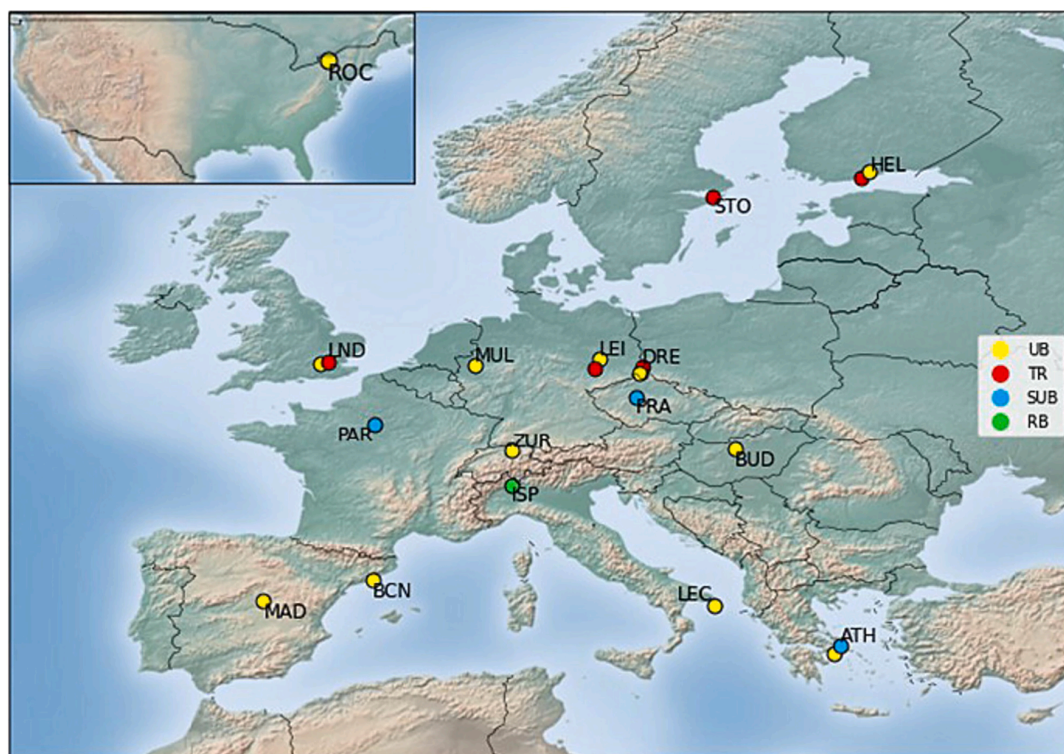


Fig. 1. Location of the cities and type of station supplying data on PNC and PNSD. UB, Urban background; TR, Traffic; SUB, Suburban background; RB, Regional background.

sites, the lower size detection limits have to be considered.

In this study, trend analyses are carried out for UFP (N_{10-100}), PNC (N_{10-800}) and the Nucleation (N), Aitken (A) and Accumulation (Ac) modes (N_{10-25} , N_{25-100} and $N_{100-800}$, respectively); and also, for the ancillary data (regulated atmospheric pollutants and meteorological parameters).

2.3. Data treatment and trend analysis

Data treatment and statistical analysis were carried out using R

statistical software (v4.2.3, [R Core Team, 2023](#)) and the *Openair* package ([Carslaw and Ropkins, 2012](#)) in R.

Data series with periods exceeding 3 years were evaluated to detect monotonic trends over the time using the non-parametric Theil-Sen method ([Sen, 1968](#); [Theil, 1992](#)). This method, robust against outliers, is commonly applied for air quality data analysis ([Carslaw and Ropkins, 2012](#)). To this end, monthly aggregation of data series was performed with a threshold of 30 % of the hourly concentration data. The selection of a 30 % threshold was based on the data availability considering the amount of data used in the study. Many sites had data availability of

Table 1

List of air quality sites supplying UFP-PNSD datasets to this study with location, type of environment and period of measurement. Coord., Coordinates; Alt., Altitude; UB, Urban Background; TR, Traffic; SUB, Suburban Background; RB, Regional Background.

City (Country)	Station Name	Station type	Acronym	Coord.; (Alt., m a.s.L.)	Period
Athens (GR)	Thissio	UB	ATH_UB	38.00 N, 23.72 E; (110)	2015–2019
Barcelona (ES)	Palau Reial	UB	BCN_UB	41.39 N, 2.13E; (80)	2013–2019
Budapest (HU)	BpART	UB	BUD_UB	47.48 N, 19.06 E; (115)	2013–2019
Dresden (DE)	Winckelmann Str.	UB	DRE_UB	51.04 N, 13.73 E; (120)	2010–2019
Helsinki (FI)	SMEAR III	UB	HEL_UB	60.12 N, 24.58 E; (26)	2009–2019
Lecce (IT)	ECO Observatory	UB	LEC_UB	40.20 N, 18.07 E; (50)	2015–2019
Leipzig (DE)	TROPOS	UB	LEI_UB	51.35 N, 12.43 E; (113)	2009–2019
London (GB)	North Kensington	UB	LND_UB	51.52 N, 0.21 W; (27)	2009–2018
Madrid (ES)	CIEMAT-Moncloa	UB	MAD_UB	40.45 N, 3.73 W; (669)	2009–2019
Mülheim an der Ruhr (DE)	Mülheim-Styrum	UB	MUL_UB	51.45 N, 6.87 E; (39)	2009–2019
Rochester NY (US)	NYS DEC	UB	ROC_UB	43.15 N, 77.55 W; (137)	2009–2019
Zurich (CH)	Kaserne	UB	ZUR_UB	47.38 N, 8.53 E; (410)	2015–2019
Dresden (DE)	North	TR	DRE_TR	51.07 N, 13.74 E; (116)	2009–2019
Helsinki (FI)	Mäkeläkatu	TR	HEL_TR	60.19 N, 24.95 E; (26)	2015–2019
Leipzig (DE)	Mitte	TR	LEI_TR	51.34 N, 12.38 E; (111)	2010–2019
London (GB)	Marylebone Rd	TR	LND_TR	51.52 N, 0.15 W; (35)	2010–2019
Stockholm (SE)	Hornsgatan	TR	STO_TR	59.32 N, 18.05 E; (20)	2010–2018
Athens (GR)	Demokritos	SUB	ATH_SUB	37.99 N, 23.82 E; (270)	2010–2019
Paris (FR)	SIRTA	SUB	PAR_SUB	48.71 N, 2.16 E; (162)	2012–2019
Prague (CZ)	Suchbát	SUB	PRA_SUB	50.13 N, 14.38 E; (277)	2012–2019
Ispra (IT)	JRC	RB	IPR_RB	45.80 N, 8.63 E; (209)	2009–2019

Table 2

Instrumentation used to measure UFP-PNSD, size range of measurement and ancillary data available in each site of the study. ND, Not-determined; SMPS, Scanning Mobility Particle Sizer; CPC, Condensation Particle Counter; MAAP, Multi-Angle Absorption Photometer; DMPS, Differential Mobility Particle Sizer; TSMPS, Twin Scanning Mobility Particle Sizer; DMA, Differential Mobility Analyser; TDMPs, Twin Differential Mobility Particle Sizer; UCPC, Ultrafine Condensation Particle Counter; OPC, Optical Particle Counter.

Station	UFP-PNSD instrumentation	Range (nm)	Ancillary data
ATH_UB	SMPS TSI 3034	10.4–487	BC, NO, NO ₂ , O ₃ , CO, T, RH, P, RAD, R
BCN_UB	SMPS TSI 3080 + CPC TSI 3772	12.2–478.3	BC, PM10, PM2.5, PM1, SO ₂ , N, NO ₂ , O ₃ , CO, T, RH, P, RAD, WS
BUD_UB	DMPS + CPC TSI 3775	10.8–816.2	PM10, SO ₂ , NO, NO ₂ , O ₃ , CO, T, RAD
DRE_UB	TROPOS-SMPS uses a Vienna-type DMA 28 cm + CPC TSI 3772	10–800	BC, PM10, SO ₂ , NO, NO ₂ , O ₃ , T, RH, P, RAD, WS, R
HEL_UB	TDMPs Hauke-type DMA 10.9 cm + CPC TSI 3025/3756 and Hauke-type DMA 28 cm + CPC 3010/3772	10–794	SO ₂ , NO, NO ₂ , O ₃ , CO, T, RH, P, RAD, WS
LEC_UB	TROPOS-SMPS uses a Vienna-type DMA 28 cm + CPC TSI 3772	10–800	BC, SO ₂ , NO, NO ₂ , O ₃ , T, RH, RAD, WS, R
LEI_UB	TROPOS-TDMPs uses Vienna-type DMAs 11 and 28 cm + CPC TSI 3025/3010	10–800	ND
LND_UB	SMPS TSI 3080 + CPC TSI 3775 with long DMA	17–604	BC, PM10, PM2.5, SO ₂ , NO, NO ₂ , O ₃ , CO, T, RH, WS
MAD_UB	SMPS TSI 3080L + CPC TSI 3775	15.1–661.2	BC, PM10, PM2.5, SO ₂ , NO, NO ₂ , O ₃ , CO, T, RH, RAD, WS, R
MUL_UB	SMPS TSI 3080 + CPC TSI 3772	14.1–495.8	PM10, PM1, NO, NO ₂ , T, RH, RAD, WS, R
ROC_UB	SMPS TSI 3071 + CPC TSI 3010	11.1–469.8	BC, PM2.5, SO ₂ , NO, NO ₂ , O ₃ , T, RH, WS
ZUR_UB	SMPS TSI 3034 + Nafion aerosol dryer	16.8–478.3	BC, PM10, SO ₂ , NO, NO ₂ , O ₃ , CO, T, RH, P, RAD, WS, R
DRE_TR	TROPOS-TSMPS uses Vienna-type DMAs 11 and 28 cm + CPC TSI 3025/3010	10–800	BC, PM10, NO, NO ₂ , O ₃ , T, RH, P, RAD, WS
HEL_TR	UHEL DMPS Vienna-type DMA + CPC Airmodus A20	11–800	BC, PM10, PM2.5, NO, NO ₂ , O ₃
LEI_TR	TROPOS-TDMPs uses Vienna-type DMAs 11 and 28 cm + CPC TSI 3025/3010	10–800	BC, PM10, SO ₂ , NO, NO ₂ , T, RH, RAD, WS, R
LND_TR	SMPS TSI 3080 + CPC TSI 3775 with long DMA	16.6–604.3	BC, PM10, PM2.5, SO ₂ , NO, NO ₂ , O ₃ , CO
STO_TR	DMPS Stockholm university handmade + CPC TSI 3010/3775	10–410	PM10, PM2.5, PM1, NO, NO ₂ , CO, T, RH, P, RAD, WS, R
ATH_SUB	SMPS TSI 3080 + CPC TSI 3772	10–550	PM10, PM2.5, NO, NO ₂ , O ₃ , T, RH, RAD, R
PAR_SUB	SMPS GRIMM 5416 + OPC GRIMM	10.9–791.5	BC, PM1, NO, NO ₂ , O ₃ , CO, T, RAD, WS
PRA_SUB	SMPS TSI 3034 rebuilt at TROPOS	10.3–519.4	PM10, PM2.5, SO ₂ , NO, NO ₂ , O ₃ , CO
IPR_RB	DMPS Vienna-type, home-made + CPC TSI 3010/3772	10–800	BC, PM10, SO ₂ , NO, NO ₂ , O ₃ , CO, RH, P, RAD, R

<50 % due to the complexity of UFP-PNSD measurements and the need of frequent instrumentation maintenance, as it is evidenced by Trechera et al. (2023). For instance, Chen et al. (2022) established a threshold of 40 %. In this study, we tried to do the trends with different thresholds of data availability and considered 30 % was the most suitable in order to have representativity of the data without losing too many months of data.

The magnitude of the trends was quantified by the Theil–Sen slope, which is the median of all the possible slopes between the data pairs and the statistical significance (ss) was also evaluated. Due to the relatively extensive temporal gaps, de-seasonalisation using Seasonal Decomposition of Time Series by Loess (Cleveland et al., 1990) was not applied. These analyses were conducted using the Openair package. It should be noted that some of the time series are relatively short for the discernment of trends, and some of the longer time series show different gradients dependent upon the time interval selected.

The individual slopes, expressed as a percentage change per year with their 95 % confidence interval, were summarised using random-effects meta-analysis due to the large heterogeneity of the data between the included sites (Chen and Peace, 2013). The mean effect was calculated for each class of site individually (urban, suburban, regional and traffic sites) as well as globally to provide a comprehensive overview of the results. Meta-analyses were carried out using the “meta” R package version 6.5–0 (Balduzzi et al., 2019).

3. Results

3.1. Pollutants related with road traffic

NO₂ decreased at all sites with a ss trend with the exception of ATH (UB and SUB), in which increasing trends were recorded (+2.64, 95 %CI = [−3.68;7.88] and +2.70, 95 %CI = [0.06;6.84] % yr^{−1}, respectively), and LEC_UB and ROC_UB, with non-ss decreasing trends (Fig. 2). The largest ss decreases were recorded for IPR_RB, HEL_TR and HEL_UB (−18.91 [−24.13;−15.00], −8.58 [−10.14;−6.85] and −6.01 [−6.97;−4.83] % yr^{−1}, respectively). Taking all the SUB sites the meta-analysis yielded a non-ss trend, while for all UB and TR sites there were marked ss decreasing trends of −3.11 [−4.01;−2.22] and −3.81 [−6.03;

−1.59] % yr^{−1}, respectively. For NO, trends were very similar to those described for the type of environment, with only a non-ss increase in LEC_UB, and ss decreases in most of the other sites (Fig. 2). In this case the meta-analysis showed ss decreasing trends for SUB, UB and TR sites (−4.34 [−6.09;−2.58], −3.75 [−4.87;−2.64] and −4.75 [−8.37;−1.14] % yr^{−1}, respectively).

BC decreased at all sites with ss trends with the exception of ATH_UB and LEC_UB, in which non-ss decreasing trends were recorded. The highest decrease again being at HEL_TR (−9.14 [−11.53;−6.80]). HEL_UB does not have a long record for this parameter (Fig. 2). The meta-analysis showed ss decreasing trends for UB and TR sites, with −4.55 [−5.84;−3.27] and −7.42 [−8.62;−6.22] % yr^{−1}, respectively. Thus, the slope increases with the approach to the TR sites.

CO decreased at all sites with ss trends with exception of BCN_UB and ZUR_UB (with non-ss decreasing trends), and BUD_UB (with a non-ss increment) (Fig. 2). The highest decrease was recorded again (as for NO₂) in IPR_RB (−19.49 [−22.77;−17.33] % yr^{−1}), followed by ATH_UB and LND_TR (−6.14 [−11.47;−0.15] and −5.37 [−5.66;−5.02] % yr^{−1}, respectively). The meta-analysis showed ss decreasing trends for SUB, UB and TR sites, with −4.74 [−5.57;−3.91] and −2.36 [−3.48;−1.23] and −4.49 [−6.27;−2.71] % yr^{−1}, respectively.

3.2. Other pollutants

PM₁₀ decreased with ss trends at 10/15 sites, and the meta-analysis yielded ss trends for SUB (−4.66 [−6.22;−3.11] % yr^{−1}), UB (−2.21 [−3.08;−1.34] % yr^{−1}) and TR (−3.81 [−4.25;−3.37] % yr^{−1}) sites (Fig. 3). Similar dominant ss decreasing trends were observed for PM_{2.5} (5/9 sites), with ss decreasing trends for the averages of SUB (−2.91 [−4.13;−1.70] % yr^{−1}), UB (−1.86 [−3.33;−0.39] % yr^{−1}) and TR (−3.72 [−5.95;−1.48] % yr^{−1}) sites (Fig. 3). For PM₁ only 4 sites provided long datasets and the meta-analysis only yielded a ss decreasing trend for IPR_RB (Fig. 3).

O₃ decreased with a ss trend only at ATH_UB (−10.14 [−12.80;−3.78] % yr^{−1}) and close to ss at BCN_UB (Fig. 4). It increased at all TR sites, with an average rate of +2.65 [0.56;4.74] % yr^{−1}; but the meta-analysis yielded non-ss trends at UB and SUB sites.

SO₂ drastically decreased in 1990–2015 (Aas et al., 2019), but it

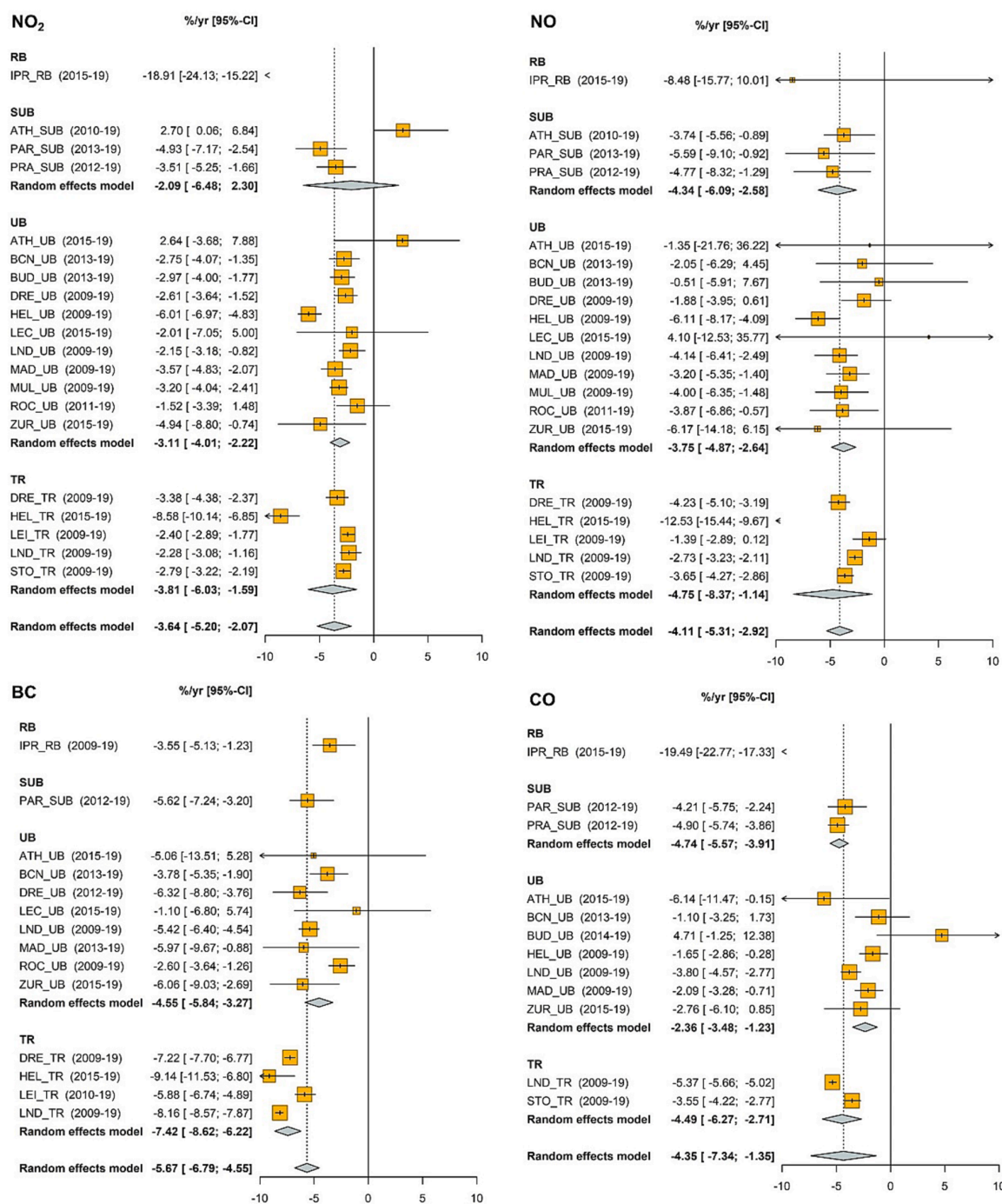


Fig. 2. Results of the trend analysis and subsequent meta-analysis for NO₂, NO, BC and CO datasets. Trends are calculated using the Theil-Sen method. Results of the meta-analysis are presented globally for each pollutant and for the different site categories (UB, TR, SUB, and RB). The dashed lined represents the global meta-analysis. Random effects model: mean effect calculated for each type of site.

continued decreasing in the analysed period. The decreases were ss at 9/13 sites (Fig. 4). The meta-analysis yielded a ss decreasing trend of $-5.59 [-8.54; -2.63] \% \text{ yr}^{-1}$, considering all sites.

3.3. Ultrafine particles

PNC decreased with ss trends at 9/21 sites (from central and north Europe, UK, USA and the regional background site in Italy) (Fig. 5). The meta-analysis yielded a non-ss trend for SUB sites, but ss for UB (-1.89

$[-3.70; 0.09] \% \text{ yr}^{-1}$), TR ($-5.13 [-7.71; -2.56] \% \text{ yr}^{-1}$) sites, and including all sites ($-2.46 [-3.89; -1.04] \% \text{ yr}^{-1}$). As it could be expected, UFP trends were similar to that of PNC, with ss decreases at 8/21 sites (the same as before but excluding HEL_TR), and only a ss increase in BUD_UB (Fig. 5). The meta-analysis again yielded a non-ss trend for the SUB sites, but in this case, for the UB sites as well. The ss decreasing trends were only seen in this case for the TR ($-4.50 [-7.41; -1.59] \% \text{ yr}^{-1}$) sites, and the global meta-analysis ($-1.98 [-3.45; 0.52] \% \text{ yr}^{-1}$). These results showed larger decreasing PNC and UFP slopes in the TR

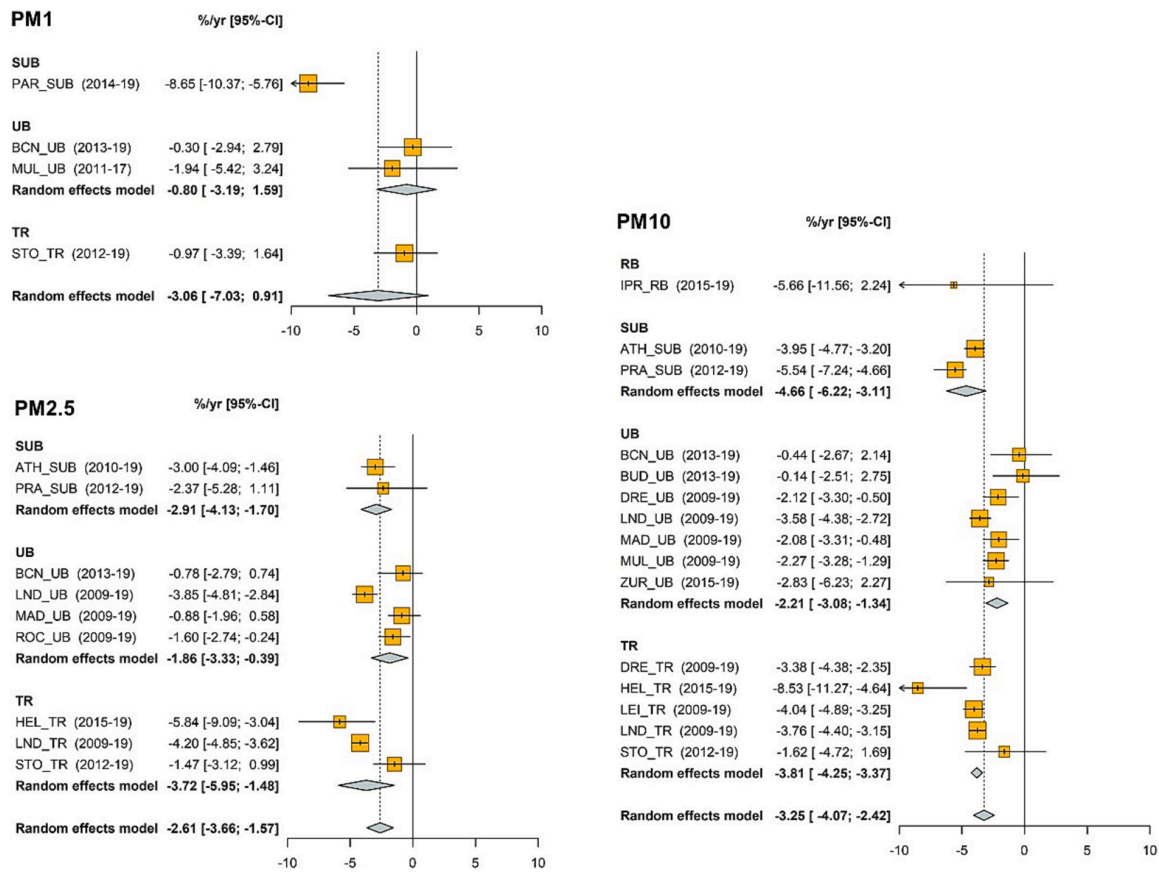


Fig. 3. Results of the trend analysis and subsequent meta-analysis for PM₁, PM_{2.5} and PM₁₀ datasets. Trends are calculated using the Theil-Sen method. Results of the meta-analysis are presented globally for each pollutant and for the different site categories (UB, TR, SUB, and RB). The dashed lined represents the global meta-analysis. Random effects model: mean effect calculated for each type of site.

sites compared with the UB ones, as also observed for the decreasing trends of the traffic-related pollutants. At IPR_RB, both PNC and UFP followed ss decreasing trends ($-1.68 [-3.01; -0.28]$ and $-1.27 [-2.04; -0.23]$ % yr⁻¹). For the UB and TR sites the slopes described above are equivalent to a decrease of -127 and -671 # cm⁻³ yr⁻¹ for PNC and -60 and -491 # cm⁻³ yr⁻¹ for UFP.

Trends for the Nucleation mode were similar to those of UFP with ss decreases at 6/21 sites (all of them from UK, central and north Europe), and ss increases at 5/21 sites (from southern and eastern Europe, but also ROC_UB, LND_UB and LEI_TR) (Fig. 6). The meta-analysis yielded non-ss decreasing trends for the SUB and TR sites and a non-ss increasing trend for UB sites. The percentage of Nucleation mode particles in PNC (N/PNC) followed ss decreasing trends at only 1/21 sites, while ss increasing trends were found at 9/21 sites. At 21 sites, non-ss increasing trends were found (Fig. 6). The meta-analysis yielded a ss decreasing trend for the SUB sites ($-2.25 [-3.62; -0.87]$ % yr⁻¹), but a positive one for the UB sites ($+3.14 [1.28; 5.00]$ % yr⁻¹). For the TR sites, only STO_TR demonstrated a non-ss decreasing trend, while the other 4 sites followed positive ones. The meta-analysis showed an increasing trend (close to the ss) for TR sites ($+1.60 [-0.31; 3.50]$ % yr⁻¹).

The Aitken mode presented dominant ss decreasing trends, with ss decreases at 11/21 sites and a ss increase only in PRA_SUB (Fig. 6). The meta-analysis yielded a non-ss trend for the SUB sites, but ss ones for UB ($-2.40 [-3.84; -0.96]$ % yr⁻¹) and TR ($-5.79 [-8.11; -3.48]$ % yr⁻¹) sites, and including all sites ($-2.81 [-4.26; -3.48]$ % yr⁻¹).

The Accumulation mode trends were similar to that of the Aitken mode, with ss decreases in 14/21 sites (Fig. 6). The meta-analysis

yielded ss decreasing trends for UB ($-3.69 [-4.98; -2.39]$ % yr⁻¹) and TR ($-5.79 [-6.83; -4.75]$ % yr⁻¹) sites, and also including all sites ($-3.92 [-4.99; -2.86]$ % yr⁻¹). IPR_RB followed also a ss decreasing trend of ($-2.39 [-4.10; -0.42]$ % yr⁻¹), but SUB sites followed a non-ss one.

3.4. Meteorological variables

Fig. S23 summarises the results of the meta-analysis for the trend analyses of T, RH, P, WS, RAD and R. The results yielded ss increasing trends for T in urban Europe with a slope of $0.13 [0.09; 0.18]$ °C yr⁻¹. In this study, 10/14 cities had ss T increases, 3/14 cities non-ss increases, and 1/15 city recorded near constant temperatures. The highest T increases were obtained for Dresden (TR site) and Leipzig with $0.33 [0.25; 0.41]$ and $0.22 [0.13; 0.31]$ °C yr⁻¹, respectively. Barcelona, Helsinki, Lecce, London, Mulheim, Paris, Stockholm and Zurich had increasing slopes of 0.13 – 0.19 °C yr⁻¹. These higher increases of T are taking place mostly in northern and central European cities, while those from southern and eastern Europe and the one from northern USA, have relatively lower slopes of increase.

The meta-analysis of the trends yielded also a ss slight increase of RAD, with $0.65 [0.15; 1.15]$ % yr⁻¹, without major regional differentiations. The other four meteorological parameters evaluated did not provide ss trends and in most cases, the slopes were close to 0.

3.5. Analysis by city

The analysis by city is shown in Supplementary Figs. S1–S22 and

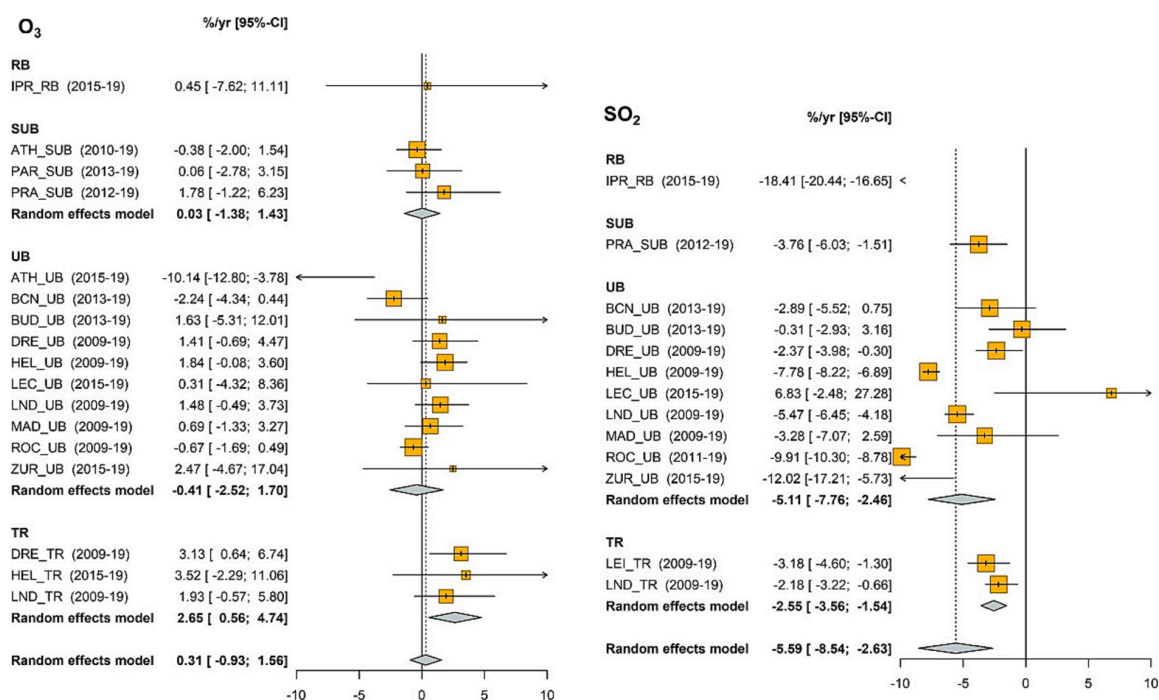


Fig. 4. Results of the trend analysis and subsequent meta-analysis for O₃ and SO₂ datasets. Trends are calculated using the Theil-Sen method. Results of the meta-analysis are presented globally for each pollutant and for the different site categories (UB, TR, SUB, and RB). The dashed lined represents the global meta-analysis. Random effects model: mean effect calculated for each type of site.

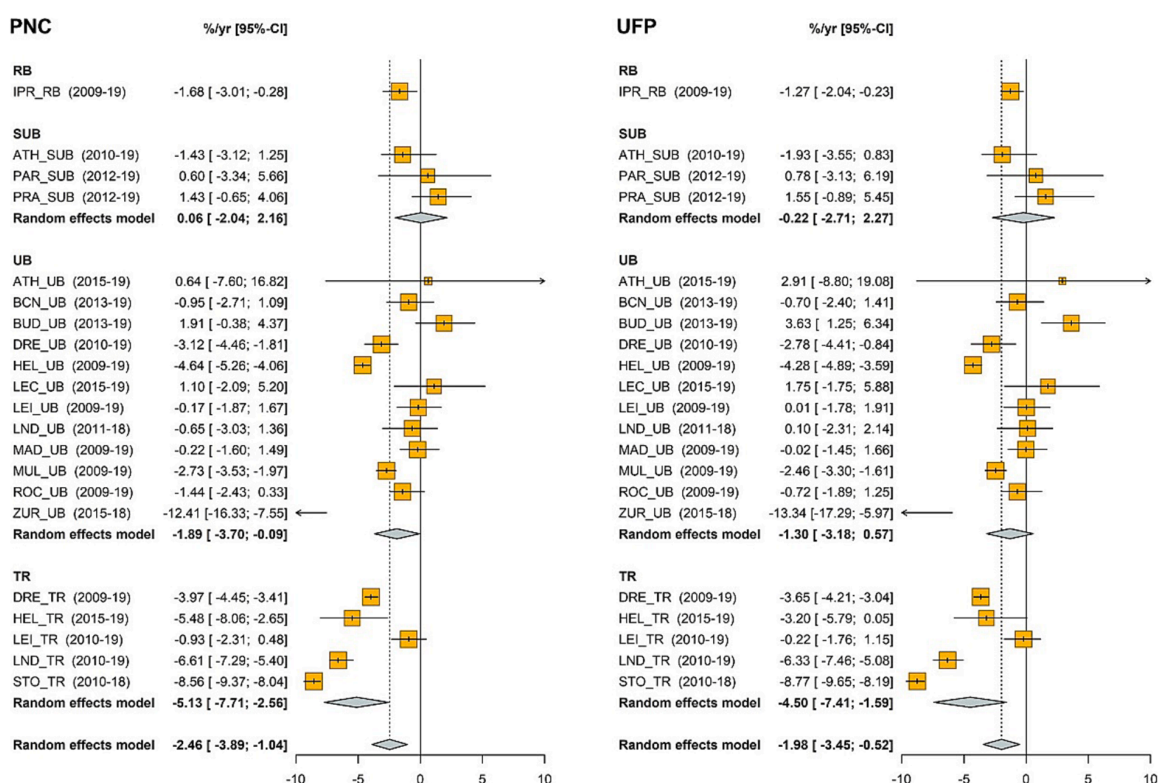


Fig. 5. Results of the trend analysis and subsequent meta-analysis for PNC and UFP datasets. Trends are calculated using the Theil-Sen method. Results of the meta-analysis are presented globally for each pollutant and for the different site categories (UB, TR, SUB, and RB). The dashed lined represents the global meta-analysis. Random effects model: mean effect calculated for each type of site.

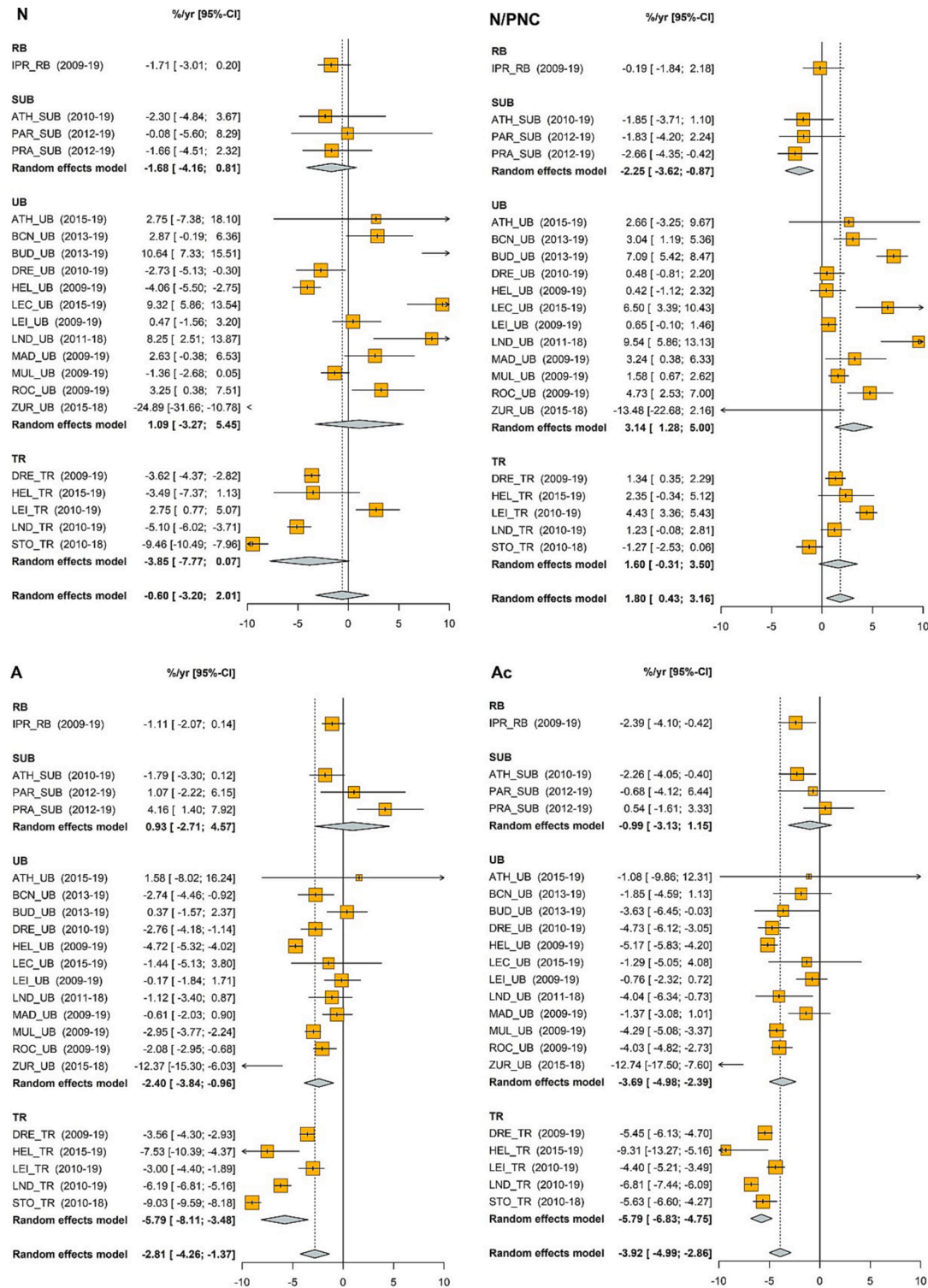


Fig. 6. Results of the trend analysis and subsequent meta-analysis for Nucleation mode (N), percentage of Nucleation mode particles in PNC (N/PNC), Aitken mode (A) and Accumulation mode (Ac) datasets. Trends are calculated using the Theil-Sen method. Results of the meta-analysis are presented globally for each pollutant and for the different site categories (UB, TR, SUB, and RB). The dashed lined represents the global meta-analysis. Random effects model: mean effect calculated for each type of site.

interpreted in [Supplementary text T1](#).

4. Discussion

4.1. Trends of NO, NO₂, BC, CO, PM, O₃ and SO₂

Very similar NO₂ declines were found at all site types, consistent with the lower emissions of NO_x from EURO 6/VI diesel vehicles. For NO, there was also a decreasing trend at the vast majority of cities, consistent with NO₂ concentrations and declining emissions of traffic-related NO_x. Trends in NO were generally steeper than trends in NO₂ (21 % and 25 % higher for NO for UB and TR sites). This steeper NO decrease might be attributed to the progressively lower NO/NO₂ emission rates from the diesel vehicles in the last decade ([Carslaw et al., 2016](#)). A higher-than-average downward NO₂ trend at HEL (both UB and TR sites) and an upward trend in ATH (at both UB and SUB sites) were observed, while ATH (both UB and SUB sites) showed a NO decrease. This unusual behaviour in both sites in Athens might be explained by the increase of diesel cars between 2012 and 2015, which their circulation was banned in Athens before 2012, and diesel cars are higher NO₂ emitters than gasoline cars ([Degraeuwe et al., 2017](#)). Thus, from 2012 onwards and with a gradual increase, diesel cars started contributing with primary NO₂ emissions changing the photochemical balance of NO/NO₂/O₃. LEC_UB had a NO increasing trend, but that was non-ss. Although EURO 5/V vehicles beginning in 2011 were equipped with DPF, the impact of the higher-than-expected NO_x emissions from diesel vehicles ([Anenberg et al., 2017](#)) resulted in delayed impacts of the EURO 5/V and 6/VI standards in abating ambient air NO₂ ([Grange et al., 2020](#); [Benavides et al., 2021](#)). The marked decrease of NO/NO_x ratios from vehicle emissions (according [Carslaw et al. \(2016\)](#) EURO 6 diesel cars emitted 67 % NO in NO_x, while in EURO 3 ones this proportion reached 91 %), and the delayed decrease of NO₂ from diesel vehicles (according the same study, EURO 2 diesel passenger cars emitted 0.08 g/km, while EURO 4 and early EURO 6 ones reached 0.33 and 0.14 g/km, respectively, in real world driving conditions) are probably the main causes of the differences in the slopes.

Downward ss BC trends were observed for all sites except LEC_UB and ATH_UB. These trends were consistent with the increased proportion of diesels with a DPF required by EURO 5/V ([Damayanti et al., 2023](#)). This decreasing trend was steeper than for NO₂ and NO (46 and 95 % higher than for NO₂, for UB and TR sites, respectively). LEC_UB appears to have experienced increased diesel traffic. ROC_UB (US) showed a smaller decline in BC, probably due to fewer light-duty diesel vehicles in the USA. Wood burning may also contribute to BC in some of the study sites, but [Savadkoobi et al. \(2023\)](#) reported that in most cases >75 % of the urban BC arises from road traffic. Furthermore, home heating and restaurants are additional urban sources ([Yu et al., 2019](#)).

The majority of sites followed a ss downward CO trend (BCN_UB and ZUR_UB are non-ss), probably associated with the growth of diesel share in the last decade and the lower emissions of the petrol vehicles since EURO 4 ([ICCT, 2016](#)). Thus, since 2006 (EURO 4) passenger diesel vehicles are allowed to emit only 0.50 g/km, and the petrol ones 1 g/km, while EURO 2–3 vehicles were allowed to emit 1.0–0.7 and 2.2–2.3 g/km, respectively ([ICCT, 2016](#)). These decreasing slopes were similar to those of NO₂ and NO, and were also attributed to decreased CO from road traffic. However, BUD_UB followed an upward trend similar to NO, possibly reflecting increased road vehicular activity affecting the site and might be also affected by the increased mean age of the passenger car fleet in Hungary ([Mikkonen et al., 2020](#)). ATH_UB followed a larger than average decline, suggesting a shift from gasoline to diesel vehicles, or possibly declining biomass combustion.

Downward trends were observed at all sites for PM_{2.5} (but these were non-ss for PRA_SUB, BCN_UB, MAD_UB and STO_TR). These reductions were likely the result of reduced diesel exhaust emissions, and a decline of secondary inorganic aerosol precursors ([EMEP, 2016](#), among others). The cause of the smaller reduction at BCN_UB is unknown. Among

possible causes, one possibility is the marked increase of cruise and container ship traffic ([Port de Barcelona, 2023](#)). In STO_TR, PM_{2.5} is mostly from long range transport from other countries. As expected, PM₁₀, which typically contains a high proportion of PM_{2.5}, followed a similar trend at most sites. Important contributors to PM₁₀, mostly in the PM_{2.5-10} fraction, are the non-exhaust vehicle emissions (vehicle wear) and road dust, which increased in their relative contributions at TR sites ([Amato et al., 2016](#)). Hence, most sites showed decreasing PM₁₀ trends. BCN_UB, BUD_UB, ZUR_UB and STO_TR showed limited and non-ss decreases. Possible reasons are associated with increased road traffic, and in the case of BCN, increased Saharan dust incursions or greater surface dryness facilitating road dust and soil resuspension. In the case of STO_TR, PM₁₀ could be largely associated with non-exhaust particles from road wear, then the small non-ss decreasing trend might be due to the meteorology that affects the wetness of the roads in winter.

A small increase of O₃ at the majority of sites was consistent with lower NO_x emissions, and lower O₃ titration by NO. The increasing trends might be attributed to the higher decreasing slope of NO compared with that of NO₂, which reduces the O₃ titration potential ([Colette et al., 2011](#); [Escudero et al., 2014](#); [Monks et al., 2015](#), among others). ATH_UB and BCN_UB followed decreasing O₃ trends. In ATH_UB these trends were consistent with increased NO₂, but not so in BCN_UB.

SO₂ decreasing trends were observed for most sites except LEC_UB, and were ss at most locations. These declines likely resulted from the reduced combustion of high S-fuels, such as coal and residual fuel oil ([Crippa et al., 2016](#)).

4.2. Trends of ultrafine particles and size fractions

PNC decreased with ss trends at 43 % of the sites (central and north Europe). The meta-analysis yielded a non-ss decreasing trend for SUB sites, but ss trends for UB and TR sites. For UFP, a similar pattern was found, but the ss decreasing trend was only observed at TR sites (−4.50 % yr^{−1}) and slightly lower than for PNC (−5.13 % yr^{−1}). Thus, dominant PNC and UFP decreases were observed, with higher slopes at the TR sites (171 % steeper in TR sites compared with the UB for PNC). These results suggest the effectiveness of DPF in abating the emissions and urban ambient concentrations of BC, PNC and UFP ([Damayanti et al., 2023](#)).

The Nucleation mode fraction followed very different trends among the evaluated cities. Several of them increased, most probably linked to a reduction in condensation sink potential, which facilitates new particle formation ([McMurry and Friedlander, 1979](#); [Spracklen et al., 2010](#); [Kulmala et al., 2017](#)). Others followed a decline, most notably STO_TR and LND_TR. The percentage of Nucleation mode particles in PNC (N/PNC) followed ss decreasing trends at only 5 % of the sites, while ss increasing trends were found at 43 % of the sites, and non-ss increasing trends at 29 %. At SUB sites, both Nucleation concentrations and percentage of Nucleation in PNC showed decreasing trends. These decreases might be related to a general decrease of the emission of precursors of the newly formed particles such as SO₂. However, increasing trends in N/PNC were observed for both UB and TR sites, with a 96 % higher slope in the UB sites. The higher increase of the percentage at the UB sites compared to the TR ones indicates that it probably is the decrease of the condensation sink that increased the Nucleation mode particles, more than the possible abatement effect of DPF of the finest UFP fraction, which was found to be minimal by [Damayanti et al. \(2023\)](#).

It is well accepted that close to roadways an increase in T might yield a decrease in UFP. Thus, [Weichenthal et al. \(2008\)](#) reported that each 10 °C increase in morning temperature was associated with decreases of 14560, 8160 and 11,310 #/cm³ for UFP exposures in walk, bus, and automobile environments, respectively. On the other side, photochemical nucleation might be influenced by T, RAD and RH conditions ([Hamed et al., 2011](#); [Yu et al., 2017](#); [Brines et al., 2015](#)), but also by the CS ([Tuovinen et al., 2021](#), and references therein) which might be reduced at high wind speeds. In urban environments under high insolation [Brines et al. \(2015\)](#) found that nucleation occurred with low CS,

high wind speeds, low RH and high T and insolation. However other global studies found a lower nucleation potential at higher T (Yu et al., 2017). As stated above, the only ss trends were obtained for T (increasing) and RAD (light increase), but not for RH and WS. The results showed that in 4/5 TR sites the Nucleation mode PNC decreased in cities where T increased. Thus, we cannot discard that the dominant decrease of the Nucleation mode particles at the TR sites of this study is influenced, at least partially, by an also generalised T increase. This effect is also possible for the UB sites of DRE, HEL, MUL and ZUR (northern and central Europe), were also a marked Nucleation mode decrease occurs simultaneously with a T increase. On the other hand, in the UB sites of BCN, BUD, LEC, LND, MAD and ROC, the T increase was coincident with an increase of the Nucleation mode, and accordingly an increase of photo-nucleation with T cannot be discarded. Thus, in addition of the effects of the above referred vehicles emission abatement policy actions on Nucleation mode trends, contribution from a generalised urban T increasing trends cannot be discarded. This is also applying to the generalised decreasing trends of the Aitken mode.

The concentrations of the Aitken mode fraction decreased at the majority of sites, most likely due to the previously mentioned reduced emissions of particles from diesel road traffic since EURO 5/V as reported for the street canyon of Marylebone (London) by Damayanti et al. (2023). There was, however, a ss increase at PRA_SUB. These differences are likely to have arisen due to increased road traffic or possibly wood burning.

The trends of the Accumulation mode were similar to that observed for the Aitken mode, most likely due to reduced road traffic solid mode particles due to DPF fitment. However, a non-ss positive trend at PRA_SUB was observed again.

Damayanti et al. (2023) found that for a TR site in London, the concentration of the Aitken mode (30–100 nm in their study), Accumulation mode particles, and BC decreased faster than other pollutants, and declined by -6.2 [-6.5 ; 5.8], -7.3 [-7.64 ; 6.99] and -8.3 [-8.56 ; 7.98] % yr^{-1} , respectively. These slopes were similar to the ones from our study for TR sites (-5.66 [-7.90 ; 3.42], -6.00 [-7.30 ; 4.70] and -7.48 [-8.70 ; 6.26] % yr^{-1} , respectively). They attributed these decreasing trends to the policy interventions in the road transport sector, which effectively decreased emissions of these pollutants. The 2010–2021 decreasing trends found by Damayanti et al. (2023) for the Marylebone (London) TR site are equivalent to a decrease of -242 , -643 and -328 # $\text{cm}^{-3} \text{yr}^{-1}$ for the Nucleation, Aitken and Accumulation modes, respectively. These slopes are all much higher than the ones from our study for TR sites (-144 , -380 and -141 # $\text{cm}^{-3} \text{yr}^{-1}$, respectively). However, Marylebone Road is a street canyon with $>90,000$ vehicles day^{-1} , and the effect of the traffic emission controls should be higher than at conventional TR sites. These trends are unlikely to have been substantially influenced by photochemical nucleation, which is observed on only 6 % of days at this site (Bousiotis et al., 2019), and was most notable on the wind sector carrying fresh traffic emissions to the sampler (Damayanti et al., 2023). Also, Chen et al. (2022), for a UB site in northern USA, found 2005–2020 decreases of -26 , -99 , -63 and -45 # $\text{cm}^{-3} \text{yr}^{-1}$ for the Nucleation (11–20 nm), Aitken-1 (20–50 nm), Aitken-2 (50–100 nm) and Accumulation (100–500 nm) modes, while the slopes from our study for UB sites were $+32$ (non-ss), -99 (ss) and -58 (ss) # $\text{cm}^{-3} \text{yr}^{-1}$ for the Nucleation, Aitken and Accumulation modes, respectively.

According to the aforementioned study, diesel vehicles in Europe played a predominant role in exhaust PM traffic contributing to ambient PM_{10} and $\text{PM}_{2.5}$ concentrations, primarily due to the PM emissions limits for diesel compared to gasoline vehicles. This distinction persisted until the implementation of EURO 5/V and 6/VI standards in 2011 and 2015, respectively, which mandated the use of DPF.

Diesel PM is largely BC, with a major mode at 40–80 nm (Hopke et al., 2022), and a second finer mode of liquid particles from condensed engine oil vapour (Kittelson et al., 2006; Harrison et al., 2018) formed by nucleation of semi-volatile organic compounds (SVOCs) as emissions

are diluted and cooled (Charron and Harrison, 2003).

More recently, Giechaskiel et al. (2022) demonstrated that the solid PNC emissions of EURO 6 diesel passenger cars are low due to the implementation of DPF. However, periodically, the trapped particles are oxidised (active regeneration), and the solid PNC emissions during this regeneration are only partly covered by the regulations. Furthermore, the PNC of volatile particles are also not included in the regulations and can be extremely high. Giechaskiel et al. (2022) measured the PNC emissions simultaneously at the tailpipe and the dilution tunnel and found that the weighted (i.e., considering the emissions during regeneration) solid PNC did not exceed the specific limit of 6×10^{11} # km^{-1} for $N_{>23}$. However, the weighted volatile PNC emissions were many orders of magnitude higher, up to 3×10^{13} # km^{-1} . Furthermore, it was found that the volatile PNC were strongly affected by desorption phenomena and that the high volatile PNC during regenerations even interfered with the 10 nm solid PNC measurements at the dilution tunnel, thus having a high impact in the Nucleation mode emissions.

At present, the EU regulates PNC emissions from vehicles only for those >23 nm. Very recently, Lintusaari et al. (2023) showed that the <23 nm particles dominated the non-volatile number concentrations and that the traffic emissions of non-volatile <10 nm particles can be even 3 times higher than those >10 nm. Yet, only a fraction of urban <10 nm particles consisted of non-volatiles.

Accordingly, the emissions of vehicular PM mass decreased markedly since 2011, followed with even larger decreases since 2016 when the implementation of EURO 6/VI required a removal efficiency of 99.9 % of all solid, carbonaceous emissions of UFP (Calderón-Garcidueñas and Ayala, 2022). Furthermore, many of the study cities implemented the Low Emission Zones (LEZ), which have been found to reduce BC concentrations (Holman et al., 2015; Baldauf et al., 2016). However, according to Damayanti et al. (2023) the decrease of emissions of Nucleation mode particles during real-world urban driving has been negatively affected by the nucleation of SVOCs that are currently not removed by emission controls. Since the formation of Nucleation mode particles occurs in the diluting exhaust gases, the process is inherently variable, depending upon the dilution ratio and environmental conditions. These factors might account for different trends found for the Nucleation mode in the study cities. Furthermore, the variability of the frequency and intensity of regional (photo) nucleation from precursors emitted from other sources than traffic, such as SO_2 from combustion sources, or of other sources of Nucleation mode particles (industry, shipping or airports) might cause also different trends among cities (Junkermann et al., 2011, 2018; Brines et al., 2015; Salma et al., 2016, 2019; Harrison et al., 2019; Rivas et al., 2020; Trechera et al., 2023).

5. Conclusions

This study evaluates long-term trends (5–11 years in 2009–2019) of ultrafine particle (UFP) concentrations and different particle size modes (UFP (N_{10-100}), PNC (N_{10-800}) and the Nucleation, Aitken and Accumulation modes (N_{10-25} , N_{25-100} and $N_{100-800}$, respectively)), based on particle number size distribution (PNSD) data from 21 sites (12 from urban background, 5 from traffic sites, 3 from suburban background and 1 from regional background) from 15 European and 1 US cities. Additionally, the trends in other pollutants and meteorological variables were evaluated in order to support interpretations.

UFP concentrations in urban Europe are largely influenced by road traffic emissions (Trechera et al., 2023, and references therein) as are other pollutants such as NO_x (i.e., NO and NO_2), CO, and BC. Concentrations of SO_2 , BC, $\text{PM}_{2.5}$ and PM_{10} are influenced by other sources. In most cases European air quality policies resulted in marked reductions of emissions of these pollutants (EEA, 2023).

The results of this study show that in most studied European cities clear abatement of the BC, NO_2 , PM, and the Aitken and Accumulation mode particle concentrations followed the implementation of diesel particle filters (DPFs), from 2011 (EURO 5/V), and the subsequent

abatement of NO_x emissions due to the controls required by EURO 6 and VI standards, which came into force in 2015, among other road traffic policies. Other air quality policy measures also generally produced SO₂ and CO decreases.

Reductions in urban NO_x emissions are expected to lead to an increase in O₃ due to a lesser titration by NO and to the reduction of NO_x concentrations in a VOCs-limited O₃ formation regime. This overall behaviour was observed at the majority of sites, especially at the traffic sites. Given the fall in NO₂ and SO₂ concentrations, reduced nitrate and sulphate are also to be expected, and accordingly of PM.

The high influence of the road traffic emissions in the UFP, PNC, BC and NO₂ urban concentrations is also demonstrated by the higher declining slopes reached at the traffic (TR) sites compared to the urban background (UB) ones. Thus, these were 141, 57, 63 and 23 % higher for the Aitken mode particles, Accumulation mode particles, BC and NO₂, respectively.

The trends in the Nucleation mode particles were far more diverse (ss decreases at 6/21 sites and increases at 5/21 sites), with a non-ss increasing trend obtained for the UB sites. However, downward non-ss trends were obtained for the TR and SUB sites. The reduction of the Nucleation mode particles at TR sites was 50, 50, 11 and 93 % lower than those obtained for Aitken and Accumulation mode particles, NO_x and BC, respectively. This is most probably due to inefficient removal of the semi-volatile diesel particle fraction by DPFs and a contribution from gasoline vehicles, expected to be predominantly in this size range, as previously reported by [Chen et al. \(2022\)](#) and [Damayanti et al. \(2023\)](#) for specific sites from US and Europe. These varying trends in the Nucleation mode particle number concentration are also affecting the UFP and total PNC trends, because of the high proportion of the Nucleation mode particles in both concentration ranges. Also the 9-fold decrease of BC versus NO_x is probably result of the efficient impact of EURO 5 DPFs in abating ambient BC and the negative effect of the 'diesel gate' on ambient NO_x.

The diverse trends obtained for the Nucleation mode particles in UB sites might be due not only to the lack of emission controls for semi-volatile organic compounds escaping from DPFs, but also due to a reduction in condensation sink potential, which facilitates new particle formation. Moreover, at some sites, the presence of other substantial lower-mode UFP sources, such as photochemical nucleation, industry, shipping and aircraft emissions, also influence the trend of the Nucleation mode.

Moreover, the urban T increasing trends might have also influenced those of PNC, Nucleation and Aitken modes. The results showed that for some sites the decrease of the Nucleation mode particles might be influenced by a T increase, but also that in other sites an increase of photo-nucleation might be associated to a T increase.

Overall, the results presented here demonstrate the positive effects of the air quality policies implemented in Europe in abating air pollutants, including the Aitken and Accumulation mode particles, but less for the Nucleation mode. The different modes of the PNSD should be evaluated independently to assess further policies promulgated to abate urban UFPs.

CRedit authorship contribution statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

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4.2.3 Paper III

Source apportionment of ultrafine particles in urban Europe

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ABSTRACT

There is a body of evidence that ultrafine particles (UFP, those with diameters ≤ 100 nm) might have significant impacts on health. Accordingly, identifying sources of UFP is essential to develop abatement policies. This study focuses on urban Europe, and aims at identifying sources and quantifying their contributions to particle number size distribution (PNSD) using receptor modelling (Positive Matrix Factorization, PMF), and evaluating long-term trends of these source contributions using the non-parametric Theil-Sen's method. Datasets evaluated include 14 urban background (UB), 5 traffic (TR), 4 suburban background (SUB), and 1 regional background (RB) sites, covering 18 European and 1 USA cities, over the period, when available, from 2009 to 2019. Ten factors were identified (4 road traffic factors, photonucleation, urban background, domestic heating, 2 regional factors and long-distance transport), with road traffic being the primary contributor at all UB and TR sites (56–95 %), and photonucleation being also significant in many cities. The trends analyses showed a notable decrease in traffic-related UFP ambient concentrations, with statistically significant decreasing trends for the total traffic-related factors of -5.40 and -2.15 % yr^{-1} for the TR and UB sites, respectively. This abatement is most probably due to the implementation of European emissions standards, particularly after the introduction of diesel particle filters (DPFs) in 2011. However, DPFs do not retain nucleated particles generated during the dilution of diesel exhaust semi-volatile organic compounds (SVOCs). Trends in photonucleation were more diverse, influenced by a reduction in the condensation sink potential facilitating new particle formation (NPF) or by a decrease in the emissions of UFP precursors. The decrease of primary PM emissions and precursors of UFP also contributed to the reduction of urban and regional background sources.

1. Introduction

Particulate matter (PM) is recognized as a major air pollutant due to its well-established harmful effects on human health. Evidence points to its association with respiratory and cardiovascular diseases, among others, as well as increased mortality (WHO, 2021a). These health impacts are influenced both by the size and composition of the particles. Ultrafine particles, defined as particles with diameters less than 100 nm, contribute a very small fraction to the PM mass concentrations (measured in terms of mass per unit volume, $\mu\text{g m}^{-3}$) but represent a substantial portion of particle number concentrations (PNC, measured in particles per cubic centimetre, $\# \text{cm}^{-3}$). In urban environments, UFPs contribute to 84 ± 3.7 % of total PNC, based on data from 27 studies across Europe (Trechera et al., 2023). Consequently, PNC is frequently utilized as a surrogate for UFP. Particle number size distributions (PNSD) are measured by calculating PNCs for specific size ranges. PNSDs categorize particle into three size modes: Nucleation (<25 nm), Aitken (25–100 nm) and Accumulation (100–1000 nm) (Seinfeld and Pandis, 2016; CEN, 2020; ISO, 2023). It is important to highlight that ambient UFP concentrations are not regulated by the EU's ambient air quality standards, while PM is (2008/50/CE, EC, 2008). However, the draft for a new proposal of an EU air quality directive (EC, 2023) requires both UFP and PNSD measurements to be implemented in routine air quality monitoring supersites, together with black carbon (BC), ammonia (NH_3), and other advanced air quality parameters.

Despite the increasing attention on UFPs, the epidemiological results regarding their health effects are often inconsistent (Ohlwein et al., 2019; US EPA, 2019; WHO, 2021b). Cassee et al. (2019) and Rivas et al. (2021) reported that inconsistency in short-term health effects studies might be due to the lack of standardization in UFP measurement methods, limited long-term data available for analysis, the high variability of UFP concentrations in both space and time, the limited representation of human exposure to UFPs resulting from the use of only one monitoring station per city in most studies, and the different sources contributing to UFP concentrations in each region. While the most recent WHO Air Quality Guidelines did not establish specific guidelines for UFP or PNC, they recommend more extensive, harmonised

measurements to assess whether establishing specific guidelines is warranted (WHO, 2021b). CEN (2020), ACTRIS (2021), RI-URBANS (2022) and Trechera et al. (2023) have suggested that total PNC should be measured with a lower size detection limit of 10 nm, and PNSDs should be assessed over a range of 10 to 800 nm.

Identifying the UFP sources and their contributions is essential to develop cost-effective abatement policies, since several studies suggest that negative health effects may be enhanced with decreasing particle size. Thus, the small size of UFPs allows them to penetrate deep into the respiratory system, potentially translocating through the alveoli and affecting various organs (Oberdörster et al., 2005; Peters et al., 2006; Kreyling et al., 2014; Salma et al., 2015; Cassee et al., 2019). This behaviour has been linked to an increased risk of premature mortality (Wichmann et al., 2000; Ibaldo-Mulli et al., 2004; Tobías et al., 2018; Schwarz et al., 2023).

Typically, receptor models for source apportionment use chemical composition data to provide information on sources of PM mass concentrations (Amato and Hopke, 2012; Pancras et al., 2013; Amato et al., 2016; Zíková et al., 2016; Taghvaei et al., 2018). However, sources dominating PNC differ from those dominating particle PM mass concentrations, thus a number of studies applied receptor modelling for the source apportionment of UFP-PNSD, using the variability of the PNSD instead of the chemical composition (Liu et al., 2014; Brines et al., 2014; Beddows et al., 2015; Sowlat et al., 2016; Rivas et al., 2019; Vörösmarty et al., 2024; Xu et al., 2024). Hopke et al. (2022) identified 55 peer-review articles reporting source apportionment of PNSD in 102 locations/time periods. Almost all of these studies were performed with Positive Matrix Factorization (PMF, Paatero and Tapper, 1994), the most frequently used receptor modelling method (Ogulei et al., 2007; Liu et al., 2014; Beddows et al., 2015; Rivas et al., 2019; Squizzato et al., 2019; Hopke et al., 2023). However, other methodologies were also used such as Principal Component Analysis (PCA, Wehner and Wiedensohler, 2003; Chan and Mozurkewich, 2007; Oliveira et al., 2009; Pey et al., 2009; Cusack et al., 2013; Khan et al., 2015) and k-means clustering (Beddows et al., 2009; Dall'Osto et al., 2012, 2019; Wegner et al., 2012; Brines et al., 2014, 2015; Chen et al., 2021). Rodríguez and Cuevas (2007) also developed a methodology to split PNCs affected by fresh

vehicle exhaust emissions into two components: N1, accounting for primary particles directly emitted in the particle phase or nucleating immediately after the emissions, and N2, accounting for the new particle formation (NPF) and other secondary UFP. Casquero-Vera et al. (2021) refined this method to estimate N1 from traffic and biomass burning sources.

Studies on source apportionment of UFPs in urban areas identify road traffic as the main source (Zhou et al., 2005; Pey et al., 2009; Dall'Osto et al., 2012; Harrison et al., 2011; Kumar et al., 2014; Brines et al., 2015; Beddows and Harrison, 2019; Rivas et al., 2019; Hopke et al., 2022, 2024; Kalkavouras et al., 2024), typically accounting for > 70 % of the annual UFP emissions (Hopke et al., 2022). Particles emitted from vehicles can be formed in the engine or in the atmosphere after emission from the tailpipe (Charron and Harrison, 2003; Kumar et al., 2014; Rönkkö and Timonen, 2019; Damayanti et al., 2023). Two traffic sources are usually reported with two major number modes with peaks around 30–35 nm, commonly ascribed to spark-ignition (gasoline) vehicles, and 60–80 nm, associated with diesel vehicle emissions (Ogulei et al., 2007; Liu et al., 2014; Hopke et al., 2022; Vörösmarty et al., 2024). It is also suggested that the smaller mode particles represent freshly traffic particles emitted by vehicles on nearby roads (Gu et al., 2011), or that can be associated to nucleation of particles generated during dilution of diesel exhaust semi-volatile organic compounds (SVOCs) (Harrison et al., 2011; Damayanti et al., 2023). The larger mode is also associated with the coagulation of the particles moving away from the sources, i.e., aged traffic emission (Zhu et al., 2002; Gu et al., 2011). However, traffic exhaust emissions also result in particles smaller than 30 nm in diameter, both as primary emissions (Rönkkö et al., 2017) and due to the nucleation of the particles immediately after the emission to the atmosphere as they dilute and cool, leading to delayed primary particles (Harrison et al., 2011; Trechera et al., 2023).

Besides traffic, other sources contribute to UFP. Photochemical nucleation lead to NPF events, characterised by PNSD peaking at the finest sizes. These events are enhanced by high insolation and wind speed, low relative humidity, low pre-existing particle surface area (i.e., low condensation sink) and available precursor gases such as SO₂ (Kulmala and Kerminen, 2008; Brines et al., 2015; Trechera et al., 2023). Emissions from harbours and airports also typically prevail in the Nucleation mode (Westerdahl et al., 2008; Keuken et al., 2015; Rivas et al., 2019; Stacey et al., 2020). Other sources identified contributing to urban UFP concentrations are characterised by modes close to or coarser than 100 nm, such as urban background (with high load from traffic), domestic heating, regional background and long-range transport.

There are only a few available studies analysing long-term trends of UFP (Simon et al., 2020; Mikkonen et al., 2020; Presto et al., 2021; Chen et al., 2022; Damayanti et al., 2023; Garcia-Marlès et al., 2024). Garcia-Marlès et al. (2024) evaluated 2009–2019 trends of UFP concentrations and different particle size modes, based on PNSD measurements from 21 sites from 15 European and 1 USA cities. The results showed significant decreases in the Aitken and Accumulation modes, suggesting a positive impact of the implementation of EURO 5/V and 6/VI vehicle standards on European air quality and the growing use of Diesel Particle Filters (DPF), but inconsistent results in the Nucleation mode particles due to insufficient controls. Additionally, long-term trends in the contribution of UFP sources can also be analysed. However, most of the studies on source apportionment of UFP did not cover periods long enough to perform trend analyses of source-specific PNCs. Recently, Hopke et al. (2024) conducted the source apportionment of PNSD from a site in USA (Rochester, New York) covering a long period and analysed the trends of the sources.

The project RI-URBANS (Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial Areas) is a European research initiative funded by the European Union's Horizon 2020 program (grant 101036245). Its primary goal is to develop tools for the measurement and analysis of advanced air quality parameters, with the aim of enhancing pollution control, improving air

quality policies, and assessing health impacts. A key focus of RI-URBANS is the measurement and application of UFP-PNSD in air quality assessments. Prior RI-URBANS studies analysed UFP in urban Europe. Trechera et al. (2023) reported the phenomenology of UFP and compared 2017–2019 concentrations across different cities and environments, Liu et al. (2023) calculated and evaluated aerosol Lung Deposited Surface Area (LDSA) concentrations, and Garcia-Marlès et al. (2024) focused on the long-term trends of UFP. In line with these, the present study aims to identify and quantify sources contributing to UFP-PNSD using the receptor model PMF, and to assess long-term trends of the source contributions. The source apportionment of 24 datasets is conducted for a period covering up to 11 years (2009–2019), when available.

2. Methodology

2.1. Monitoring sites and instrumentation

This study analyses 24 PNSD datasets collected between 2009 and 2019, with data coverage ranging from 3 to 11 years. These data were compiled by the RI-URBANS project and supplied from advanced air quality monitoring networks and research sites, and already used with other purposes in previous studies by Trechera et al. (2023), Liu et al. (2023) and Garcia-Marlès et al. (2024). The data covers 24 sites across 18 European cities and one in the USA, with different climate and urban structure patterns, UFP concentrations and sources. These sites include (see Fig. 1 and Table 1):

- Fourteen urban background (UB) sites: Antwerp (ANT_UB), Athens (ATH_UB), Barcelona (BCN_UB), Budapest (BUD_UB), Dresden (DRE_UB), Granada (GRA_UB), Helsinki (HEL_UB), Lecce (LEC_UB), Leipzig (LEI_UB), London (LND_UB), Madrid (MAD_UB), Mülheim (MUL_UB), Zurich (ZUR_UB), and Rochester (ROC_UB) in New York State in USA.
- Five traffic (TR) sites: Dresden (DRE_TR), Helsinki (HEL_TR), Leipzig (LEI_TR), London (LND_TR) and Stockholm (STO_TR).
- Four suburban background (SUB) sites: Athens (ATH_SUB), Lille (LIL_SUB), Paris (PAR_SUB) and Prague (PRA_SUB).
- One regional background (RB) site: Ispra (IPR_RB).

Hourly PNSD averages were used in this study, with data restricted to 2019 to avoid the influence of pollution reductions caused by COVID19-lockdowns, as noted in several studies (Salma et al., 2020; Dinoi et al., 2021; Eleftheriadis et al., 2021; Petit et al., 2021; Putaud et al., 2021, 2023).

In addition to the PNSD data, ancillary pollutants (including BC, NO₂, NO, CO, SO₂, O₃, PM₁₀, PM_{2.5} and PM₁) were collected where available (see Table 1). These measurements were taken either from the same monitoring locations or nearby air quality monitoring sites, by instrumentation fulfilling European standards. PNSD data used in this analysis is publicly accessible through the EBAS database (<https://ebas.nilu.no/>).

PNSD datasets were collected using different types and models of Mobility Particle Size Spectrometer (MPSS), as detailed in Table S1. Although ACTRIS (2021) and CEN (2020) guidelines recommend a measurement size range of 10–800 nm, some instruments had coarser lower detection limits, starting above the recommended 10 nm. Furthermore, drastic concentration jumps were detected in the lower sizes in some datasets, thus these ranges were removed. Consequently, 50 % of the sites (BCN_UB, BUD_UB, GRA_UB, LND_UB, MAD_UB, MUL_UB, ROC_UB, ZUR_UB, HEL_TR, LND_TR, LIL_SUB, PAR_SUB) started measurement between 11 and 20 nm. As stated by Trechera et al. (2023), the lower size detection significantly affects the total PNC, thus this variation must be accounted for when comparing sources with PNC prevalence in the Nucleation mode.

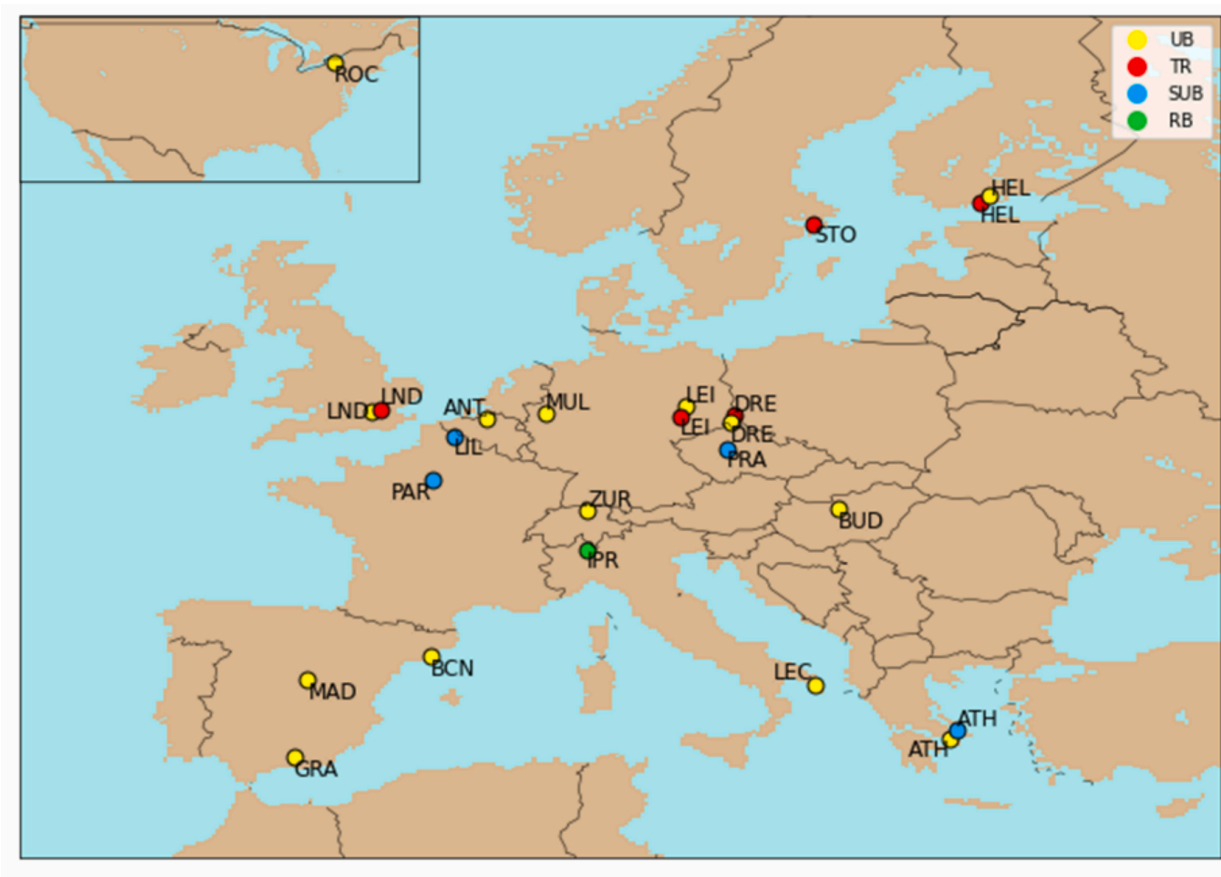


Fig. 1. Map showing the location of the cities and type of stations providing data on particle number concentration (PNC) and particle number size distribution (PNSD). Station categories include Urban background (UB), Traffic (TR), Suburban background (SUB), and Regional background (RB).

Table 1
List of air quality monitoring sites providing UFP-PNSD datasets for this study, including measurement periods, size range (in nm), and available ancillary pollutants. Categories include Urban Background (UB), Traffic (TR), Suburban Background (SUB), and Regional Background (RB); ND indicated not determined.

City (Country)	Station Name	Acronym	Period	Range	Ancillary pollutants
Antwerp (BE)	Borgerhout	ANT_UB	2017–2019	10–808	BC, PM ₁₀ , PM _{2.5} , SO ₂ , NO, NO ₂ , O ₃
Athens (GR)	Thissio	ATH_UB	2015–2019	10–470	BC, PM ₁₀ , NO, NO ₂ , O ₃ , CO
Barcelona (ES)	Palau Reial	BCN_UB	2013–2019	12–478	BC, PM ₁₀ , PM _{2.5} , SO ₂ , NO, NO ₂ , O ₃ , CO
Budapest (HU)	BpART Lab	BUD_UB	2009–2019	11–816	PM ₁₀ , SO ₂ , NO, NO ₂ , O ₃ , CO
Dresden (DE)	Winckelmann Str.	DRE_UB	2010–2019	10–800	PM ₁₀ , SO ₂ , NO, NO ₂ , O ₃
Granada (ES)	UGR	GRA_UB	2017–2019	11–496	BC, PM ₁₀ , SO ₂ , NO, NO ₂ , O ₃ , CO
Helsinki (FI)	SMEAR III	HEL_UB	2009–2019	10–708	SO ₂ , NO, NO ₂ , O ₃ , CO
Lecce (IT)	ECO Observatory	LEC_UB	2015–2019	10–800	BC, SO ₂ , NO, NO ₃ , O ₃
Leipzig (DE)	TROPOS	LEI_UB	2009–2019	10–800	ND
London (GB)	North Kensington	LND_UB	2011–2018	17–604	BC, PM _{2.5} , SO ₂ , NO, NO ₂ , O ₃ , CO
Madrid (ES)	CIEMAT-Moncloa	MAD_UB	2013–2019	15–661	BC, PM ₁₀ , SO ₂ , NO, NO ₂ , O ₃ , CO
Mülheim an der Ruhr (DE)	Mülheim-Styrum	MUL_UB	2009–2019	14–496	PM ₁₀ , NO, NO ₂
Rochester NY (US)	NYS DEC	ROC_UB	2011–2019	12–470	BC, PM _{2.5} , SO ₂ , NO, NO ₂ , O ₃
Zurich (CH)	Kaserne	ZUR_UB	2015–2019	17–478	BC, PM ₁₀ , SO ₂ , NO, NO ₂ , O ₃ , CO
Dresden (DE)	North	DRE_TR	2009–2019	10–800	BC, PM ₁₀ , NO, NO ₂ , O ₃
Helsinki (FI)	Mäkeläkatu	HEL_TR	2015–2019	11–800	BC, PM ₁₀ , PM _{2.5} , SO ₂ , NO, NO ₂ , O ₃
Leipzig (DE)	Mitte	LEI_TR	2010–2019	10–800	BC, PM ₁₀ , SO ₂ , NO, NO ₂
London (GB)	Marylebone Rd	LND_TR	2010–2019	17–604	BC, PM _{2.5} , SO ₂ , NO, NO ₂ , O ₃ , CO
Stockholm (SE)	Hornsgatan	STO_TR	2012–2018	10–410	PM ₁₀ , PM _{2.5} , NO, NO ₂ , CO
Athens (GR)	Demokritos	ATH_SUB	2010–2019	10–550	PM ₁₀ , NO, NO ₂ , O ₃
Lille (FR)	Villeneuve d'Ascq	LIL_SUB	2017–2019	20–496	PM ₁₀ , NO, NO ₂ , O ₃
Paris (FR)	SIRTA	PAR_SUB	2013–2019	11–800	BC, PM ₁ , NO, NO ₂ , O ₃
Prague (CZ)	Suchbát	PRA_SUB	2012–2019	10–475	PM ₁₀ , PM _{2.5} , SO ₂ , NO, NO ₂ , O ₃ , CO
Ispra (IT)	JRC	IPR_RB	2016–2019	10–800	BC, PM ₁₀ , SO ₂ , NO, NO ₂ , O ₃ , CO

2.2. Positive matrix factorization

Positive Matrix Factorization (PMF; [Paatero and Tapper, 1994](#)) is the most recently used data analysis method to identify and apportion the sources of PNSD. It is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into two matrices: factor contributions and factor profiles. These need to be interpreted to identify the source types using measured source profile information, and emissions inventories.

The increased use of PMF came as a result of the freely available implementation provided by the U.S. Environmental Protection Agency (US EPA; [Norris et al., 2014](#)). However, the current implementation of USEPA PMF 5.0 program, a graphic user interface coupled to the underlying solver, the multilinear engine 2 (ME-2; [Paatero, 1999](#)), does not handle datasets of more than approximately 500,000 data points. Thus, [Hopke et al. \(2023\)](#) developed a tool that would permit the PMF analysis of large datasets using ME-2, which is used in this study. A roadmap of the process of the PMF followed in this study is presented in [Fig. 2](#).

The datasets included hourly averaged PNSD data combined with hourly concentrations of the ancillary pollutants available at each site, to help the identification of the sources. PMF requires individual uncertainty estimates for each data value. Uncertainties in the PNC values were estimated using the approach of [Ogulei et al. \(2007\)](#), [Squizzato et al. \(2019\)](#) and [Hopke et al. \(2024\)](#), calculated by:

$s_{ij} = \alpha_{ij} + C_3 \cdot n_{ij}$ where $\alpha_{ij} = 0.01(n_{ij} + \bar{n}_{ij})$ and C_3 is a constant determined by trial-and-error testing values between 0.001 and 0.15. The

lowest and highest bins of the PNSD have been reported to have increased measurement error ([Wiedensohler et al., 2018](#)). Consequently, uncertainties for the 3 % lowest and 3 % highest size bins were multiplied by a factor of 2, and by a factor of 1.5 for the subsequent 3 % lower and 3 % higher size bins ([Rivas et al., 2019](#)).

Uncertainties for the ancillary pollutants were calculated following the methodology established by [Polissar et al. \(1998\)](#). An additional uncertainty K was applied to all the variables. The values of C_3 and K that optimise the model are presented in [Table S2](#).

The datasets for each site were independently analysed by PMF. PMF was run multiple times for different numbers of factors (sources), from 3 to 7 factors. Then, the number of factors was finally determined examining the results and choosing the best solution. These best solutions were selected based on the accepted criteria and guidelines ([Belis et al., 2019](#); [Hopke et al., 2023](#)), considering: (i) scaled residuals approximately randomly distributed between -3 and 3 , (ii) a $Q_{\text{true}}/Q_{\text{exp}}$ ratio close to 1, (iii) profile uncertainties determined by the displacement (DISP) method, and (iv) the provision of the most physically meaningful profiles and temporal behaviours.

To support the identification of the sources, the contributions of each factor to the variance of the co-located ancillary pollutants, daily patterns, seasonality and polar plots were evaluated. The data analysis and plots were performed with the R statistical software (v4.2.3, [R Core Team, 2023](#)) and the package *Openair* ([Carslaw and Ropkins, 2012](#)).

2.3. Splitting of Nucleation into Photonucleation and traffic sources

At some sites, sources with prevalence in the Nucleation mode included particles generated by photonucleation processes, and those emitted from road traffic; and PMF was not able to separate them into two factors due to the similar PNSDs. Thus, these sources were split following a methodology developed by [Rodríguez and Cuevas \(2007\)](#) and [Rivas et al. \(2019\)](#). NO_x and BC were used as proxies for traffic emissions (see [Table S3](#)). Considering that the morning concentration peak of this source is built mostly by primary traffic particles, NO_x or BC concentrations were multiplied by a scaling factor so it matched the PNC of this source at morning peak hour. This scaling factor (the ratio between the PNC of the source and the proxy) was calculated for each day. The percentile 10 % of the PNC of this source/BC or NO_x at morning traffic rush hours at each site was selected as a scaling factor. The selection of the 10th percentile was based on previous studies ([Rodríguez and Cuevas, 2007](#); [Reche et al., 2011](#); [Kulmala et al., 2016](#); [Kalogridis et al., 2018](#); [Nie et al., 2022](#)) and the variability of PNC of each year of data in this study. Also, at some sites, all the particles from photonucleation were attributed to traffic sources during the night hours, as no photonucleation would be expected since these particles are formed due to solar radiation. Examples of the splitting (DRE_UB and MUL_UB) are shown in [Figures S1 and S2](#).

2.4. Trend analysis

Inter-annual trend analysis of the source contributions was conducted using the same methodology as described by [Garcia-Marlès et al. \(2024\)](#). For detection of monotonic trends, datasets covering at least 4 complete years within a period of more than 5 years were analysed using the non-parametric Theil-Sen method ([Sen, 1968](#); [Theil, 1992](#)), a widely used approach for air quality trend assessments ([Carslaw and Ropkins, 2012](#)). Monthly averages were calculated with a threshold of 30 % of the hourly concentration data. The Theil-Sen slope, which represents the median of all the possible slopes between data pairs, was used to quantify the magnitude of the trends, while statistical significance (ss) was evaluated using the *Openair* R package ([Carslaw and Ropkins, 2012](#)).

A random effects meta-analysis was performed on the estimated trends for each source, using individual slopes expressed as percentage changes per year along with their 95 % confidence intervals. The mean

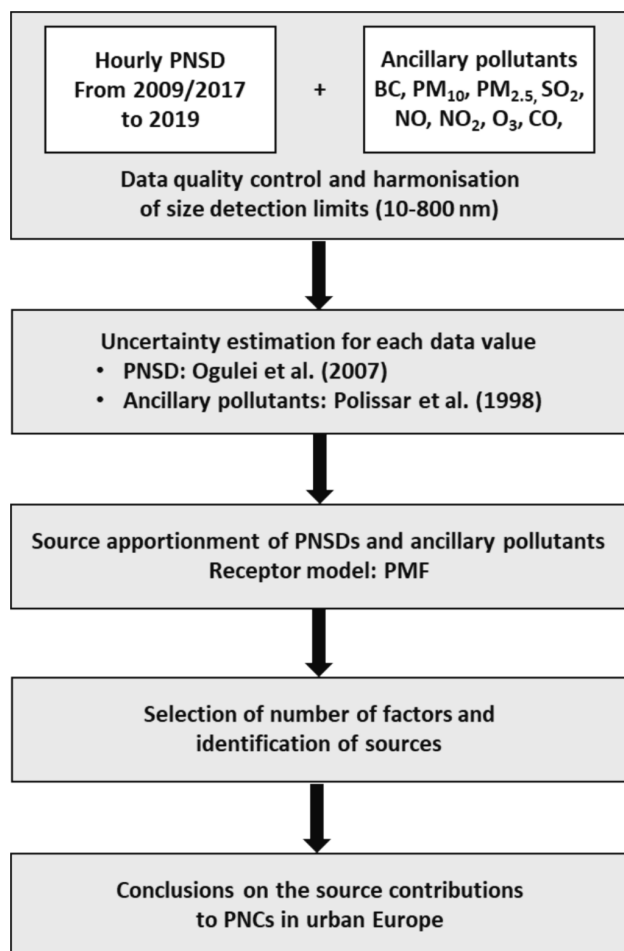


Fig. 2. Roadmap of the process followed for the source apportionment of PNSD using PMF.

effect was calculated separately for site type (urban, suburban, and traffic) as well as for all sites combined. The analysis was conducted using the *meta* R package (Balduzzi et al., 2019).

3. Results and discussion

3.1. Sources identified

From 4 to 7 factors of PNC were identified at each site. In total, 10 different factors were found associated to different aerosol sources for the 24 datasets of the study: *Traffic-1*, *Traffic-2*, *Mixed traffic*, *Traffic-Nucleation*, *Photonucleation*, *Urban background*, *Domestic heating*, *Regional-1*, *Regional-2* and *Long-distance transport*. However, in some cases a general nucleation factor (<25 nm) obtained by PMF was further split into *Photonucleation* and *Traffic-Nucleation* (see Table S3). In other cases, a coarser factor (25–35 nm) associated with traffic was split into *Traffic-1* and *Photonucleation*. The *Photonucleation* source was then added to the existing *Photonucleation* source obtained directly from PMF at the site. This additional source is linked to particles generated by photochemical processes but with a regional origin, as evidenced by the coarser size mode and its peaks at midday, albeit with a delay compared to a local photonucleation.

The sources identified at each site are characterised by different PNSDs and trends. Modes' sizes with the highest PNC for the different sources at each site are reported in Table 2. These modes are affected by the size ranges of each site, thus the lower detection limit has to be considered, especially in the nucleation sources. Results of the source apportionment for each site are interpreted in Supplementary text T1 and presented in Figures S3-S77 and Tables S4-S27. These show the profiles for PNSD, the contributions to the variance of ancillary pollutants, daily, monthly and weekly trends of PNC, and polar plots.

3.1.1. Traffic sources

Four different factors associated with road traffic source were identified at the sites of this study, with similar temporal behaviours but different PNSDs (Table 2; Fig. 3). *Traffic-1* and *Traffic-2* are typically documented in the existing literature (Hopke et al., 2022).

Traffic-1 identified in this study is characterised by major size modes with peaks ranging between 23 and 39 nm (Table 2). This size mode

associated to traffic is commonly attributed to gasoline vehicle emissions in ambient air urban PNSD measurements (Ogulei et al., 2007; Liu et al., 2014; Hopke et al., 2022, 2024; Vörösmarty et al., 2024), freshly emitted traffic particles on nearby roads (Gu et al., 2011), or nucleation of particles generated during dilution of diesel exhaust semi-volatile organic compounds (SVOCs) (Harrison et al., 2011, Damayanti et al., 2023). *Traffic-2* is characterised by size mode peaks at 55–119 nm (Table 2), and is associated with primary diesel vehicle emissions (Ogulei et al., 2007; Liu et al., 2014; Hopke et al., 2022). Some studies also suggest that this larger traffic mode is due to the coagulation of the particles moving from the sources, i.e., an aged traffic emission (Zhu et al., 2002; Gu et al., 2011). The traffic origin of both factors is supported by peaks observed during rush hours in the morning and evening, lower values in summer in cities where the car fleet decreases on holidays, and low weekends PNCs (Fig. 3). Also, these have mainly a local origin and predominate at low wind speeds (Supplementary text T1). However, a few cities, such as Antwerp, Leipzig, Paris, Rochester or Stockholm, have higher total PNC in summer, and most of the source contributions are also higher in summer, probably due to meteorological conditions favouring more stagnation in summer (Hopke et al., 2024). *Traffic-2* profiles exhibit higher contributions to the variance of BC and NO_x compared to *Traffic-1* (Supplementary text T1). This aligns with interpretations, as diesel exhaust emits BC-rich soot particles when not equipped with diesel particle filters (DPFs) (Damayanti et al., 2023, and references therein) and is responsible for most of the traffic-related NO_x emissions (He et al., 2015). Both profiles also contribute to CO, with *Traffic-2* additionally contributing to PM_x.

Mixed traffic is characterised by major size mode peaks at 43–55 nm (Table 2), falling within the range between *Traffic-1* and *Traffic-2*. It represents a highly traffic-influenced urban background; thus, it is mixed with other sources, keeping the daily, seasonal and weekly patterns of traffic sources (Fig. 3). This factor also predominates at low wind speeds and has contributions to the variance of BC, NO_x and CO (Supplementary text T1). *Traffic-1* and *Traffic-2* were well separated and identified at all the UB and TR sites except for ATH_UB, BUD_UB and both sites in Helsinki (HEL_UB and HEL_TR). These factors were also identified in ATH_SUB and PAR_SUB, but not at the other SUB and RB sites. The sites where these factors were not individually identified presented the *Mixed traffic* factor, but the three distinct traffic factors

Table 2

Size modes with the highest PNC of the sources identified in each site. In parentheses, size mode of lower peaks in the PNSD. TR-Nucl, Traffic-Nucleation; TR-1, Traffic-1; Mixed TR, Mixed traffic; TR-2, Traffic-2; Photonuc., Photonucleation; UB, Urban background; Dom heat, Domestic heating; Reg-1, Regional-1; Reg-2, Regional-2; LDT, Long-distance transport.

Station	TR-Nucl.	TR-1	Mixed TR	TR-2	Photonuc.	UB	Dom heat	Reg-1	Reg-2	LDT
ANT_UB	11	39		83	11			271 (30)	191 (10)	
ATH_UB		28	47		16	160 (28)	84			
BCN_UB		27	43	88	13				151 (31)	300 (69, 13)
BUD_UB	20		52		20	114		250 (114, 39)	169 (16)	
DRE_UB		28	54	119	11			291	232 (48)	
GRA_UB	13	23	43	98	13			175 (27)		
HEL_UB	16		45					200 (28)	80	
LEC_UB	11	27		60	11		106	260	48 (208)	
LEI_UB	13	28	54	106	13			364 (95, 14)	232	
LND_UB		37		87	22			255 (65)		
MAD_UB	19	37		69	19	188 (24)			151 (20)	
MUL_UB	15	29	50	88	15	188 (36)		322 (78, 19)		
ROC_UB	18	35		67	18		41	246 (20)	249	
ZUR_UB		31		55	20		106	300 (31)	188 (32)	
DRE_TR	16	38		85	16			260 (28)	208 (28)	
HEL_TR	13		55					213 (11)	30 (176)	
LEI_TR	14	39		95	14			291 (25)		
LND_TR	19	34		81				87 (255)		
STO_TR	17	35		79				198		
ATH_SUB		34		70	12				178 (25)	
LIL_SUB	24		51		24	106		269 (74)	46 (188)	
PAR_SUB		28		74				199	104	
PRA_SUB			45		16		82	195 (32)		
IPR_RB			45		18		99	198	36 (198)	

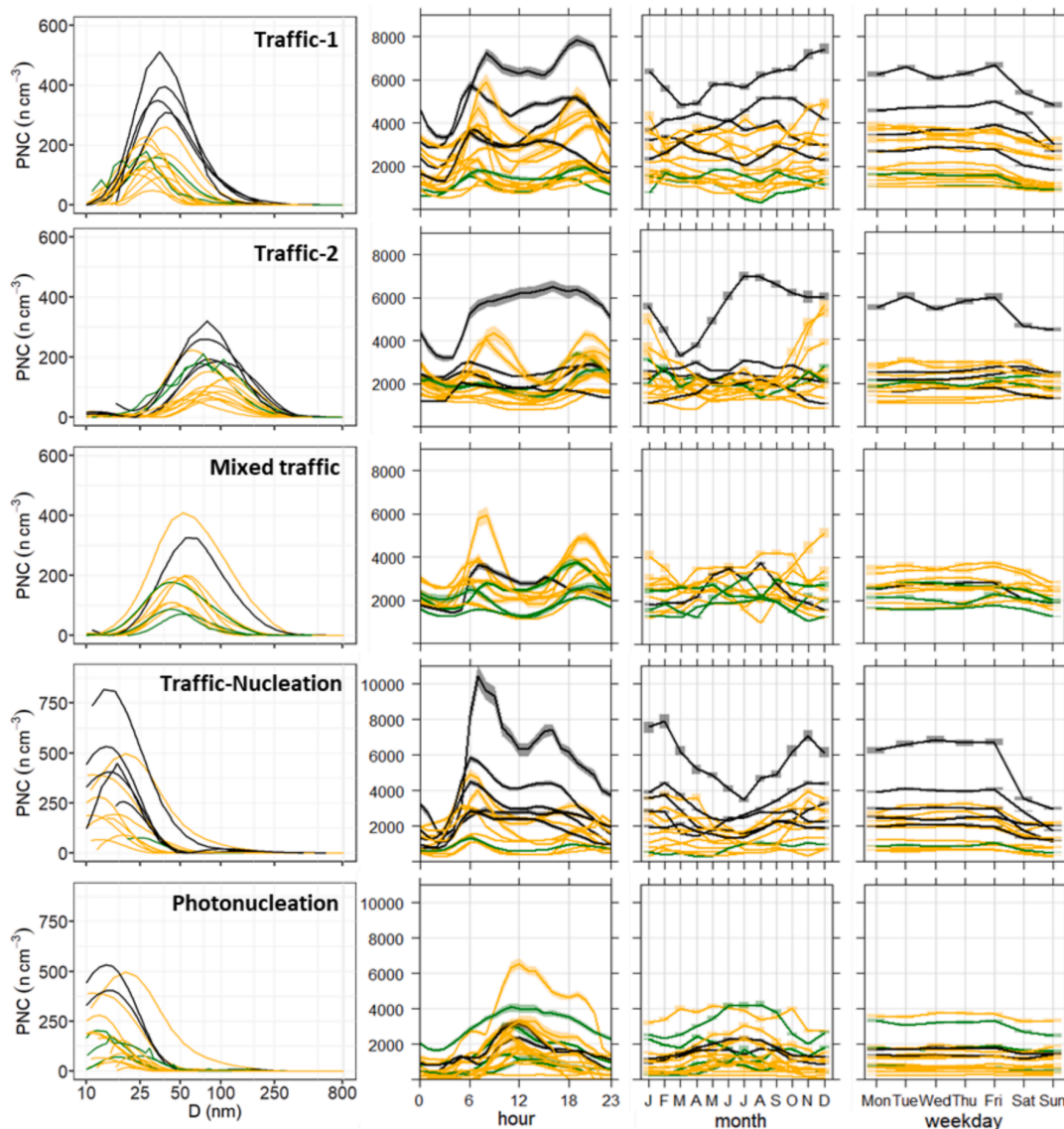


Fig. 3. Particle number size distribution (PNSD) profiles and daily, monthly and weekly patterns of *Traffic-1*, *Traffic-2*, *Mixed traffic*, *Traffic-Nucleation*, and *Photonucleation* sources across all sites. Urban background (UB) sites are denoted in yellow, Traffic (TR) sites in black, and Suburban background (SUB) and Regional background (RB) sites in green. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

were identified at several UB sites (BCN_UB, DRE_UB, GRA_UB, LEI_UB and MUL_UB). The absence of separate identification of *Traffic-1* and *Traffic-2* at some sites may be attributed to aerosol dynamical processes affecting ultrafine particles during transport and air mass recirculation, which results in the mixing of particles from different origins (Hamed et al., 2007; Junkermann and Hacker, 2018; Lv et al., 2020). Due to the collinearity of temporal patterns in particles emitted by road traffic, PMF struggles to distinguish these two sources, especially at suburban sites.

Traffic-Nucleation is characterised by size mode peaks at 11–20 nm (Table 2), falling into the lowest size bins. The differences in modes between the sites are linked to the lower size detection limit, affecting the Nucleation mode PNC. This traffic exhaust factor is associated to the nucleation of the particles immediately after the emission of SVOCs (from a mixture of those arising from the regeneration of DPF in diesel vehicles and from catalytic converters in gasoline ones) to the atmosphere as they dilute and cool, leading to NPF (Harrison et al., 2011; Mamakos and Martini, 2011; Trechera et al., 2023; Saarikoski et al.,

2024). It also follows the daily, seasonal and weekly patterns of traffic sources (Fig. 3) and predominates at low wind speeds (Supplementary text T1). This factor was identified at most sites after applying the methodology to split the factor from *Photonucleation* (see methodology) (ANT_UB, BUD_UB, GRA_UB, LEC_UB, LEI_UB, MAD_UB, MUL_UB, ROC_UB, DRE_TR, LEI_TR and LIL_SUB). At other sites, it was identified directly from PMF (HEL_UB, HEL_TR, LND_TR and STO_TR). Furthermore, it has been shown that *Traffic-Nucleation* increases with decreasing ambient temperature. Weichenthal et al. (2008) found a decrease of around 10,000 # cm⁻³ per each increase of 10 °C, at sites close to traffic emissions.

3.1.2. Photonucleation

Photonucleation is characterised in this study by size modes with peaks ranging between 11 and 22 nm (Table 2) and represents most of the PNCs in the Nucleation mode at urban sites. As mentioned above, the lower size detection limit influences the profiles at each site. This source is associated with photochemically induced NPF events, which are

enhanced by high insolation (Kulmala and Kerminen, 2008; Brines et al., 2015). Thus, peaks at midday/late morning are observed across all sites, and PNCs are higher in summer, coinciding with peak solar radiation (Fig. 3). At certain sites, BCN_UB as an example (see Supplementary text T1), smaller peaks during traffic rush hours in the morning and evening persist. This result suggests that the source still includes Nucleation mode particles from traffic exhaust even after applying the methodology to distinguish photonucleation particles from traffic nucleation particles. While traffic-related pollutants are not prominent in the *Photonucleation* source, elevated levels of SO₂ and O₃ are observed at most sites (Supplementary text T1). This source was identified at all sites except for HEL_UB, HEL_TR, LND_TR, STO_TR and PAR_SUB. However, for the latter, photonucleation particles are present in the source identified as *Regional-2*.

Photonucleation particles are formed in high-insolation and high wind

speed environments, leading to NPF events (Kulmala and Kerminen, 2008; Brines et al., 2015; Salma et al., 2021; Trechera et al., 2023; among others). These events are further enhanced by a low condensation sink (McMurry and Friedlander, 1979), occurring when traffic exhaust emissions decrease, and with available SO₂. At some sites such as BCN_UB, it is mostly influenced by nucleated particles in the high SO₂ plumes emitted from the harbour, as supported by winds blowing from that direction and a previous analysis of cluster ions by Nitrate Chemical Ionisation Atmospheric Pressure interface Time of Flight Mass Spectrometer (CI-API-ToF) (Brean et al., 2020). Additionally, coal-fired power plants emitting SO₂ contribute to NPF episodes at other sites, especially in central Europe (Junkermann et al., 2011). These are generally transported at high altitudes. Insolation and associated convective dynamics lead to the growth of the mixing layer, which traps these plumes and causes Hewson-type fumigations (of these high

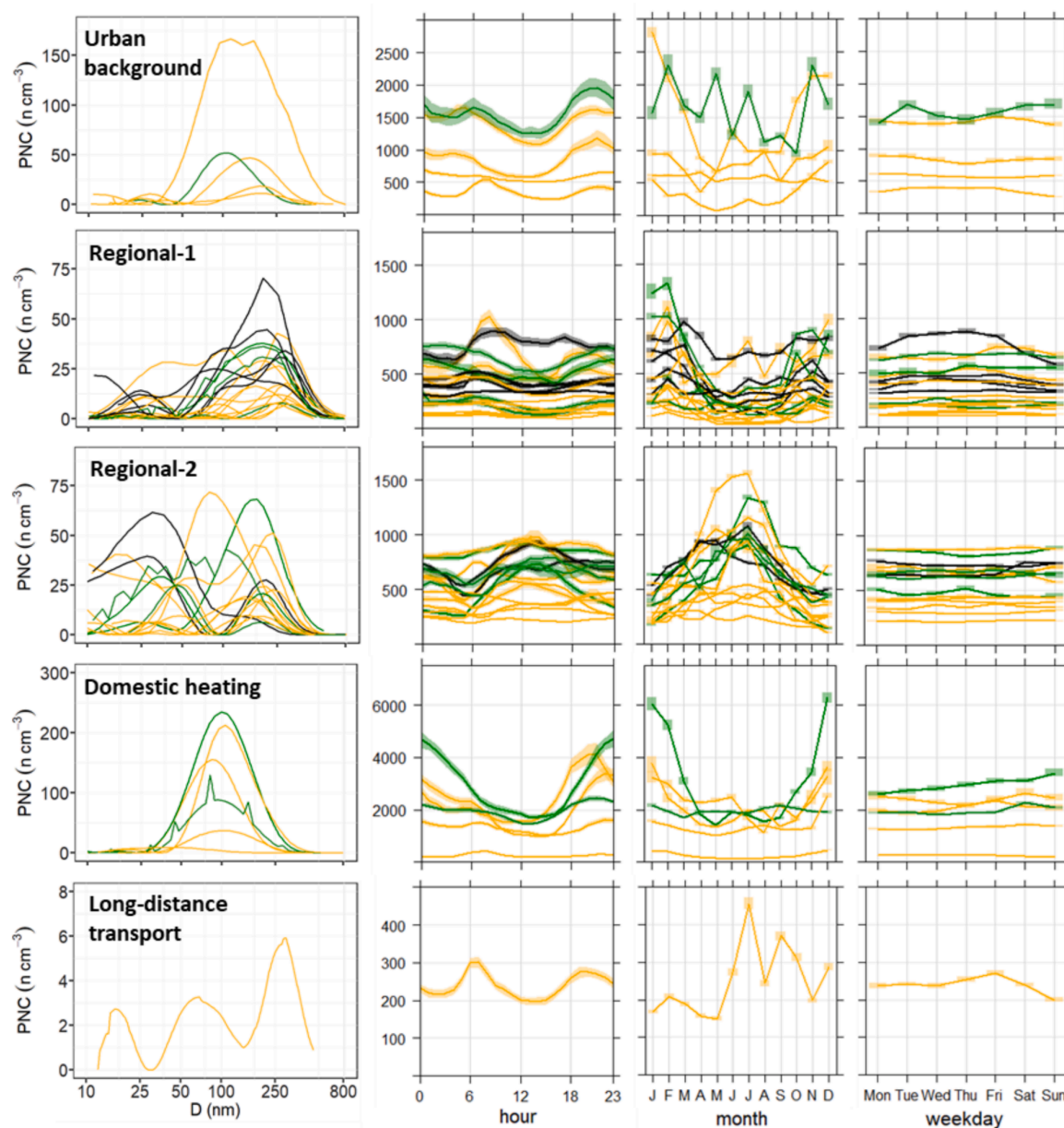


Fig. 4. Particle number size distribution (PNSD) profiles and daily, monthly and weekly patterns of *Regional-1*, *Regional-2*, *Urban background*, *Domestic heating* and *Long-distance transport* sources across all sites. Urban background (UB) sites are denoted in yellow, Traffic (TR) sites in black, and Suburban background (SUB) and Regional background (RB) sites in green. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Nucleation mode particles) on the surface (Hewson, 1955). Furthermore, regional-scale nucleation episodes might account for a high proportion of NPF (Kalkavouras et al., 2020).

3.1.3. Urban and regional background

Urban background is characterised by size modes peaks at 106–188 nm (Table 2) and follows similar traffic patterns with the highest values at rush hours and during winter (Fig. 4). It has high contributions of PM_x, BC, NO_x and CO and has a local origin (Supplementary text T1). This source, which was identified at ATH_UB, BUD_UB, MAD_UB, MUL_UB and LIL_SUB, can be described as highly traffic influenced urban background, following similar traffic profiles and patterns but with a coarser mode. It includes aged traffic particles that had grown after the emission, with a high amount of carbon agglomerates supported by the high contributions of BC (Beddows et al., 2015; Rivas et al., 2019). It is differentiated from the *Mixed-traffic* source only because of the coarser size mode of the *Urban background*.

Two different regional sources were identified due to opposite day-night and summer-winter patterns. The **Regional-1** is characterised by a multimodal distribution with main peaks ranging between 175–364 nm and secondary peaks in the Aitken and Nucleation modes (Table 2). It is higher at night and during winter (Fig. 4), and has high contributions of PM_x and traffic-related pollutants. It is associated with high wind speeds (usually easterly winds in northern and central Europe) but has also a local origin (especially in southern and eastern Europe) (Supplementary text T1). The **Regional-2** profiles also display a multimodal distribution but with finer size modes, with main peaks ranging between 80–232 nm (Table 2). It has higher PNCs at midday and during summer (Fig. 4), coinciding with higher insolation. It has high contributions of PM_x, O₃ and SO₂, and is associated with high wind speeds (Supplementary text T1). Neither of these sources exhibits a day-of-the-week pattern.

Regional-1 is mainly associated with nitrates, which are primarily formed through atmospheric oxidation of NO_x (Seinfeld and Pandis, 2016). This mainly occurs during winter, as ammonium nitrate evaporates in summer due to high temperatures, and at night, consistent with semi-volatile nitrate condensing on locally emitted particles during periods of lower temperature and higher relative humidity (Dall'Osto et al., 2009; Masiol et al., 2016). The particles in the Nucleation and Aitken mode are likely associated with the gas-to-particle conversion of local NO_x emissions whereas the Accumulation mode particles are formed during the transport and reactions of their precursor gases, NH₃ and NO_x (Kasumba et al., 2009; Squizzato et al., 2019). Furthermore, domestic emissions of UFP (higher in winter and night) might also contribute to this source.

Regional-2 is usually associated with secondary ammonium sulphate and organic aerosols. The high summer insolation (and O₃ concentrations) accelerates the oxidation of SO₂ and VOCs to produce NPF with an origin from H₂SO₄ reaction with NH₃ or amines, with a subsequent growth dominated by condensation of oxidised VOCs (Ehn et al., 2014). The ageing of these particles to build up the regional background yields a coarser size fraction. This is enhanced in the summer season, because of the higher insolation and O₃, but also biogenic VOC emissions. It has a finer PNSD than the *Regional-1* because of the high contributions of NPF followed by growth, and because of the thermal instability of ammonium nitrate in summer but not in winter. Several sites (LEC_UB, HEL_UB, HEL_TR, LIL_SUB and IPR_RB) show the main peaks of this source in the Aitken mode between 30–80 nm, along with additional peaks in the Accumulation mode. These mainly consist of nucleated particles that have grown during transport from the emission sources, especially associated with SO₂-rich plumes (Qian et al., 2007; Bousiotis et al., 2019). In the Western Mediterranean, the well-known summertime vertical recirculation of air masses (Millán et al., 2000; Gangoiti et al., 2001) is a major cause of regional O₃ pollution episodes. It also contributes to the enrichment of aerosol particles and their precursors, leading to higher levels of UFPs in *Regional-2* compared to *Regional-1*

(Carnerero et al., 2019).

3.1.4. Other sources

Domestic heating is characterised by size modes ranging between 82–106 nm in ATH_UB, LEC_UB, ZUR_UB, PRA_SUB, and IPR_RB, and a wide PNSD profile in ROC_UB, where the maximum PNC occurs at 41 nm (Table 2). It is higher at night, during winter, and at weekends (Fig. 4), when domestic burning is more frequent (Kleeman et al., 2009; Gu et al., 2011; Corsini et al., 2019), and has high contributions of BC and CO (Supplementary text T1). BC and CO are emitted as products of incomplete combustion of carbon-containing fuels, such as wood and coal (Wei et al., 2012; Chen et al., 2015). Significant emissions of UFP have been reported from these domestic combustion processes (Tiwari et al., 2014; Zhang et al., 2014; de la Sota et al., 2018; Wang et al., 2020; Kuye and Kumar, 2023). These sources include cooking and heating activities, emitted by wood-burning and pellet stoves, fireplaces, electric heaters, and kitchen stoves, among others. Additionally, PNC peaks in spring and summer could be attributed to biomass burning resulting from agricultural activities and fires (Cesari et al., 2018). However, in some sites such as GRA_UB, HEL_UB, HEL_TR, and ATH_SUB, this source was not individually identified by PMF, but contributions of domestic heating might be included in traffic sources, especially in *Traffic-2*, with higher contributions at night. For a campaign in winter 2015–2016 in GRA_UB, Casquero-Vera et al. (2021) found that primary biomass burning contributed 9 % of total PNC, but PMF was not applied.

Long-distance transport was only identified in BCN_UB and is characterised by a multimodal distribution with the main size mode at 300 nm (Table 2). Higher PNCs coincide with rush hours and during summer (Fig. 4), with high contributions of PM_x. It is mainly associated with particles from the Sahara Desert in Africa, as shown by the high contributions observed with SW winds with high velocities, and back-trajectory analyses (Supplementary text T1). A similar PNSD was found by Al-Dabbous and Kumar (2015) for a long-range transport source associated with Arabian dust events. However, lower contributions with a local origin are also observed, related to road dust emitted from traffic, explaining the diel pattern (Gu et al., 2011). Additional sources contributing might include long-range transport of polluted air masses (with high loads of sulphate and other secondary pollutants) and forest fires. Figures S12 illustrates the mean Dust Surface Concentration on days characterised by substantial dust contributions in Barcelona, coinciding with documented dust events. During these periods, elevated levels of sulphate particles (Figure S13) and emissions from forest fires (Figure S14) were also recorded. However, this source was not identified by PMF in GRA_UB, MAD_UB or LEC_UB, which might also have significant contributions from Saharan dust.

3.2. Quantitative source contributions

Fig. 5 shows the relative contribution to average PNCs of the sources identified at each study site. The sum of all the traffic contributions (*Traffic-1*, *Traffic-2*, *Mixed traffic* and *Traffic-Nucleation*) represents 70–88 % of the average PNC at 9 out of the 14 UB sites analysed, and 47–61 % at the remaining 5 sites. As it could be expected, at TR sites, traffic sources contribute even more significantly, accounting for 77–95 % of the average PNC. These contributions are higher at TR compared to UB sites in cities where PNC measurements are conducted at both types of sites. It is notable that *Traffic-Nucleation* is more relevant at TR sites (16–54 % of the average PNC at the 5 studied sites) compared to UB sites (7–38 % of the average PNC identified at 9/14 sites). Traffic nucleated particles might arise from gasoline emissions and, to a larger extent, from SVOCs escaping the DPFs in diesel vehicles (Harrison et al., 2011; Damayanti et al., 2023). These traffic particles exhibit rapid growth as they move away from the traffic source, thus at UB sites, traffic particles are generally coarser than in other traffic factors. This effect arises from coagulation and from loss of the delayed nucleated diesel traffic mode by evaporation as they move into cleaner background air (Dall'Osto

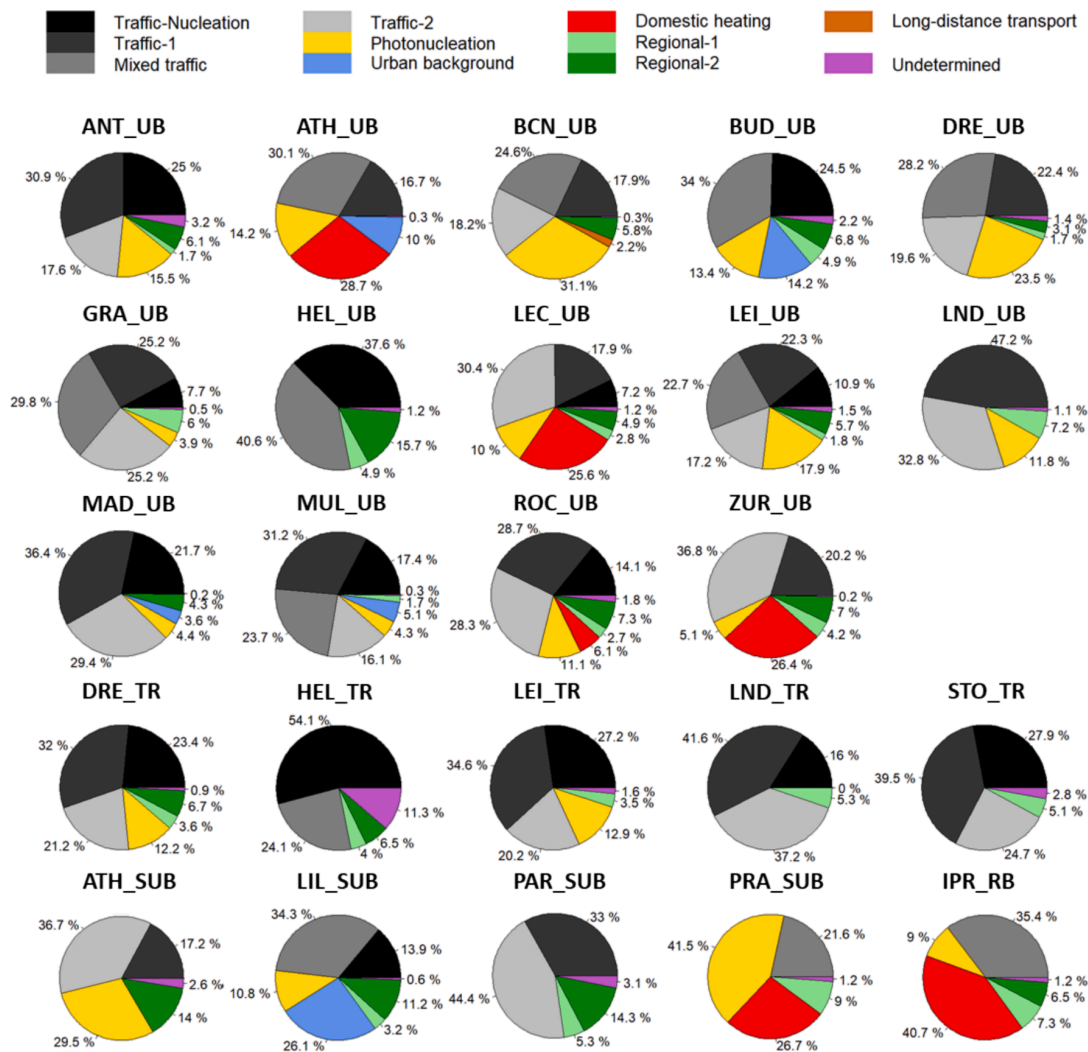


Fig. 5. Relative contributions (%) of particle number concentration (PNC) from the identified sources at each study site. The *Undetermined* category (in purple) represents the fraction of PNC not assigned to a specific source by PMF. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

et al., 2011; Harrison et al., 2016). However, it is important to note that at sites where the splitting methodology of nucleated particles was applied, the uncertainty in quantifying source contributions might be higher. Additionally, the contribution of the traffic sources may contain particles from other (less relevant) sources and be affected by the distance from the monitoring site to major roads or by specific climate/meteorological patterns of the cities. Moreover, at sites like HEL_TR, the PMF analyses resulted in regional source profiles with traffic patterns that were not possible to split into traffic/background portions, potentially leading to an underestimation of traffic contributions. At SUB and RB sites, traffic contributions are not consistently the major contributors across all locations, comprising 22–77 % of the average PNC, with PAR_SUB exhibiting the highest traffic contributions among these sites.

Accordingly, the results point to road traffic as the major contributor to UFP/PNC in urban Europe. The high contribution from traffic emissions to total PNSD has also been reported in literature for many urban environments around the world (Pey et al., 2009; Dall'Osto et al., 2012; Liu et al., 2014; Beddows et al., 2015; Brines et al., 2015; Sowlat et al., 2016; Rivas et al., 2019; Squizzato et al., 2019; Kalkavouras et al., 2024, and others).

Following traffic, *Photonucleation* is a significant contributor at

several sites, primarily driven by NPF mostly at regional scales. In BCN_UB, with relatively high insolation levels compared with other study cities (Trechera et al., 2023), this source contributes 31 % to the average PNC, notably higher during summer, but remaining significant throughout the year. As stated above, this is highly influenced by emissions of precursors from shipping. Dresden and Leipzig also exhibited substantial contributions from *Photonucleation*, accounting for 24 % (UB) and 12 % (TR) in Dresden, and 18 % (UB) and 13 % (TR) in Leipzig. Also, as stated above, these values were probably caused by surface fumigation by higher-altitude atmospheric layers (enriched in Nucleation mode particles through NPF processes) during the growth of the planetary boundary layer (Junkermann et al., 2016). This source was identified at all remaining UB sites (4–16 % of the average PNC), except for Helsinki. Additionally, it was present at all SUB and RB sites except for PAR_SUB, with the highest contribution observed at PRA_SUB. The contributions of this source may be subject to under or overestimation at sites where the nucleation splitting (traffic/photonucleation) methodology was applied.

Domestic heating contributed significantly at ATH_UB, LEC_UB, ZUR_UB, PRA_SUB, and IPR_RB (26–41 % of the average PNC) and less at ROC_UB (6 %). However, this source probably contributed also to

Regional-1, Urban background and traffic factors at sites where *Domestic heating* was not identified.

Lower contributions were observed for *Urban background* (4–14 % of the average PNC at 4/19 UB and TR sites, and 26 % at the SUB site in Lille), *Regional-1* (2–9 % at 20/24 sites), *Regional-2* (3–16 % at 16/24 sites) and *Long-distance transport* (2 % in BCN_UB). However, these contributions are also affected by other sources such as traffic.

3.3. Inter-annual trends of source contributions

Following the study by Garcia-Marlès et al. (2024), which evaluated long-term trends of UFP/PNC concentrations of most of the sites of this study, inter-annual trends of contributions of sources identified in this study were evaluated. Sites included for the trend analysis, that provide more than 4 complete years in a period longer than 5 years, are: BCN_UB, DRE_UB, HEL_UB, LEI_UB, LND_UB, MUL_UB, ROC_UB, DRE_TR, HEL_TR, LEI_TR, LND_TR, STO_TR, ATH_SUB and PRA_SUB.

Total traffic emissions (the sum of all the traffic factors) decreased with statistically significant (ss) trends at 10/14 sites, including 5/5 traffic sites and 5/6 European UB sites (Fig. 6). *Total traffic* decrease, in terms of PNC, -578 , -148 and $-42 \text{ cm}^{-3} \text{ yr}^{-1}$ for the TR, UB and SUB sites, respectively. The *meta-analysis* yielded ss trends for UB (-2.15 [-4.17 ; -0.13] % yr^{-1}), TR (-5.40 [-7.66 ; -3.13] % yr^{-1}) and all sites (-3.26 [-4.78 ; -1.75] % yr^{-1}). The highest decrease of the traffic factors was observed in *Traffic-2* (diesel), with ss decreasing trends at 9/11 sites (-4.21 [-5.60 ; -2.83] % yr^{-1} , for the global *meta-analysis*), and a non-ss decrease at 1/11 site. *Traffic-1* decreased with ss trends at 5/11 sites (-2.78 [-4.78 ; -0.78] % yr^{-1} , for the global *meta-analysis*) and with non-ss decreasing trends at 4/11 sites. *Mixed traffic* decreased with ss trends at 4/7 sites (-2.61 [-4.46 ; -0.76] % yr^{-1} , for the global *meta-analysis*) and *Traffic-Nucleation* decreased with ss trends at 4/9 sites (-2.97 [-5.77 ; -0.16] % yr^{-1} , for the global *meta-analysis*).

Thus, PNCs from traffic sources decreased at most sites (all traffic sites and all UB European sites except LEI_UB), most probably as a consequence of the implementation of emission standards on European air quality, particularly EURO 5 and 6 (for cars) and V and VI (for heavy-duty vehicles) and other local measures that might have been implemented for road traffic. The highest significant decrease for *Traffic-2* (primarily associated with diesel vehicle emissions) aligns with the requirement of EURO 5 and V for diesel vehicles to be equipped with DPF (Damayanti et al., 2023). This result is consistent with the downward trends of BC, and the Aitken and Accumulation mode particles observed by Garcia-Marlès et al. (2024). However, Chen et al. (2022) and Damayanti et al. (2023) reported that the abatement effect of DPF of the Nucleation mode particles was minimal because DPFs do not retain SVOCs emitted by diesel cars and these nucleate leading to delayed primary particles. Supporting this effect, traffic factors of finer UFP fractions, *Traffic-Nucleation* and *Traffic-1*, decreased less at most sites, and in some cases, lost the ss. Furthermore, the reduction of sulphur (S) content in diesel might have contributed to reducing UFP from road traffic. However, ultra-low-S diesel (ULSD) was implemented in 2006 in Europe and the US (Hopke et al., 2024), which was before the period covered by the datasets in this study. Other road traffic policy measures, such as Low Emission Zones (LEZ) in many cities and the Congestion Charge in some of them, also might have contributed to this reduction (Patel et al., 2023).

ROC_UB is the only site that showed a ss increasing trend for traffic sources ($+4.36$ [1.39 ; 8.36] % yr^{-1} , for *Total traffic*), with the highest increase in *Traffic-Nucleation*. Hopke et al. (2024) found compatible results for this US city. They reported a decrease in traffic-related UFP from 2006 to 2012 (attributed to the implementation of Tier 2 standards), followed by a flat evolution and an increase in 2017–2019. They suggested rise in economic activity beginning in 2017 (higher HDD vehicle traffic) as the major cause of this increase, but also a possible leak from ageing DPFs (Preble et al., 2019).

Different trends for *Photonucleation* among the sites evaluated were

observed, with ss increasing trends at 4/10 sites, non-ss trends at 6/10 (Fig. 6). The *meta-analysis* yielded a non-ss increasing trend for all sites. The increases might be linked to a reduction in condensation sink potential, which facilitates photochemical new particle formation (Spracklen et al., 2010; Kulmala et al., 2017). Trends for this source are expected to vary because of the different causes of NPF caused by photonucleation. Thus, as reported above, in some places this can be attributed to regional nucleation, in others to the impact of plumes of SO_2 -emitting coal or fuel-oil power plants, and shipping emission plumes. All these sources of precursors and atmospheric processes affect specific cities and might have had different trends in the different cities. Furthermore, there is a ss increasing trend for temperature in these cities (Garcia-Marlès et al., 2024) that might have also influenced the increase of UFP from NPF in specific regions.

Urban background decreased with ss at the only site evaluated (MUL_UB), showing a rate of -3.59 [-4.52 ; -2.53] % yr^{-1} (Fig. 7). This is consistent with the decreasing trends of traffic sources because of the influence of traffic emissions in *Urban background*.

Regional-1 significantly decreased at most of the sites, with ss decreasing trends at 7/11 sites and non-ss trends at the remaining sites. The *meta-analysis* yielded a ss decreasing trend for all sites with an average rate of -5.40 [-7.36 ; -3.43] % yr^{-1} (Fig. 7). This change is probably associated with the decrease of primary PM emissions and precursors of UFP (SO_2 , NO_x , and in less proportion VOCs and NH_3) across Europe, as reported by EEA (2023).

Regional-2 also showed a ss global decreasing trend (-1.90 [-3.56 ; -0.25] % yr^{-1}) but only decreased with ss at 2/8 sites (Fig. 7). This summer source was dominated by secondary sulphate and secondary organic aerosols where formation increased with insolation. In spite of the above observed decreased emissions of SO_2 , the lower decrease of NH_3 and VOCs, but especially the increase of O_3 in urban environments (Sicard et al., 2021) might have prevented more marked decreases of this contribution compared with the winter one. Thus, the increase of ozonolysis reactions might have yielded to an increase in the formation of secondary particles in summer. Saiz-Lopez et al. (2017) reported that in Madrid the urban increase of O_3 resulted in an increase of 60–100 % of oxidising radicals, yielding in an increase of the potential of formation of secondary inorganic and organic particles.

Domestic heating did not show global ss trends, as only two sites (ROC_UB and PRA_SUB) had enough data for the trend analysis, making it difficult to reach any conclusions. *Long-distance transport* did not show ss trend for BCN_UB.

4. Conclusions

This study identified and quantified source contributions of ultrafine particles (UFP) and evaluated long-term trends of these source contributions, based on 24 particle number size distribution (PNSD) datasets from 18 cities in Europe and 1 in the USA. These locations included 14 urban background (UB), 5 traffic (TR), 4 suburban background (SUB), and 1 regional background (RB) sites.

Ten different factors are identified contributing to UFP-PNSD (*Traffic-1*, *Traffic-2*, *Mixed traffic*, *Traffic-Nucleation*, *Photonucleation*, *Urban background*, *Domestic heating*, *Regional-1*, *Regional-2* and *Long-distance transport*), with road traffic being the major contributor at all UB and TR sites (56–95 %). *Photonucleation* is also a significant contributor at many sites (4–41 % in 16/19 cities), primarily driven by new particle formation (NPF). These cities include not only those with high insolation from southern Europe, but many from central Europe. The causes of these high contributions of NPF are diverse (shipping, plumes from combustion plants, regional nucleation).

The results showed a clear reduction in UFP traffic ambient concentrations. These trends reached statistically significant decreases for *Total traffic* (the sum of all the traffic contributions) of -5.40 and -2.15 % yr^{-1} for TR and UB sites, respectively, or, in terms of PNC, of -578 and $-148 \text{ cm}^{-3} \text{ yr}^{-1}$. This UFP abatement is most probably due to the

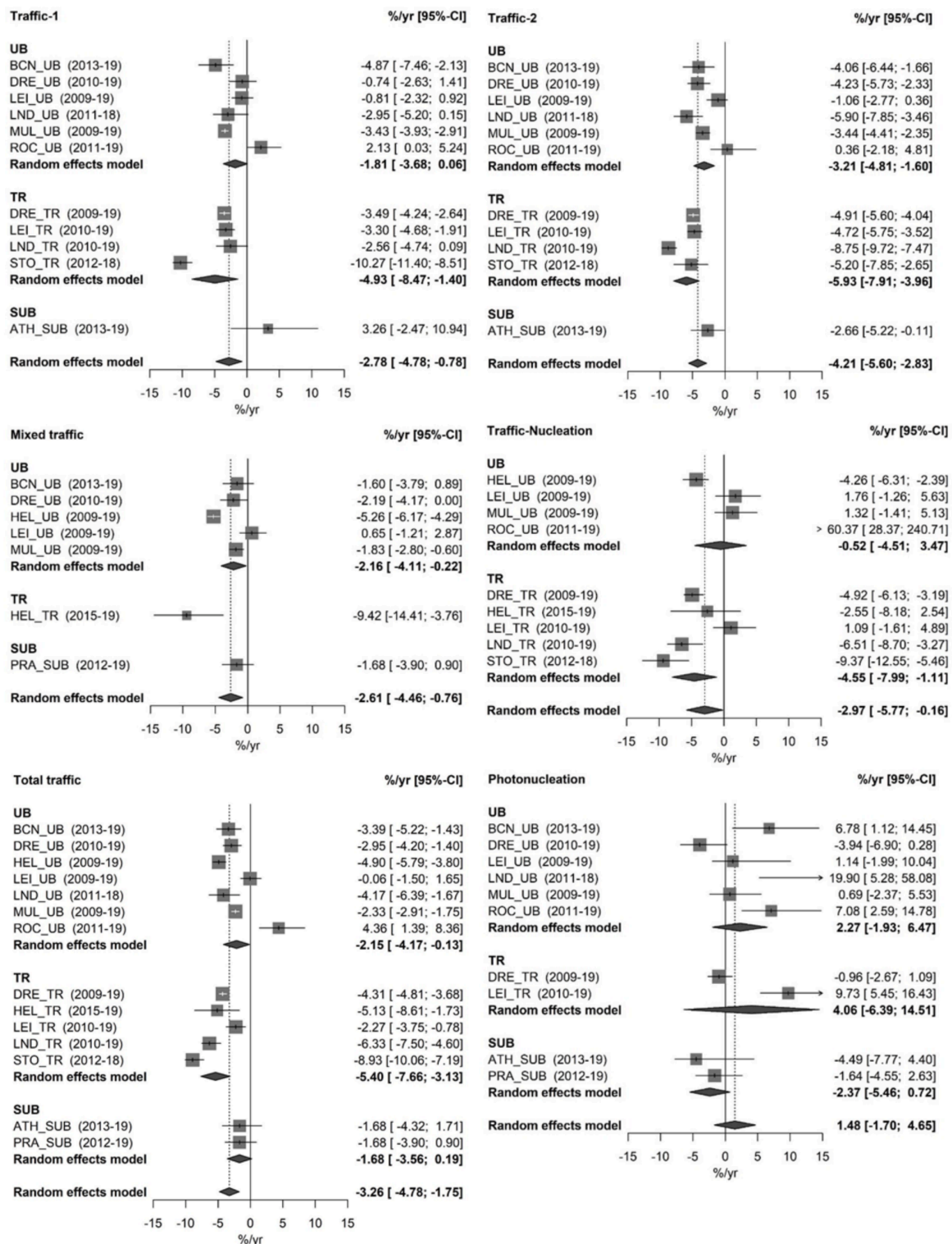


Fig. 6. Trend analysis and meta-analysis results for *Traffic-1*, *Traffic-2*, *Mixed traffic*, *Traffic-Nucleation*, *Total traffic* and *Photonucleation* sources. Trends are calculated using the Theil-Sen method. Mean effects for each site type (Urban background, UB; Traffic, TR; Suburban background, SUB; and Regional background, RB) are estimated using a random effects model. The meta-analysis results are presented for each pollutant across the different site types, as well as for global trends (dashed line).

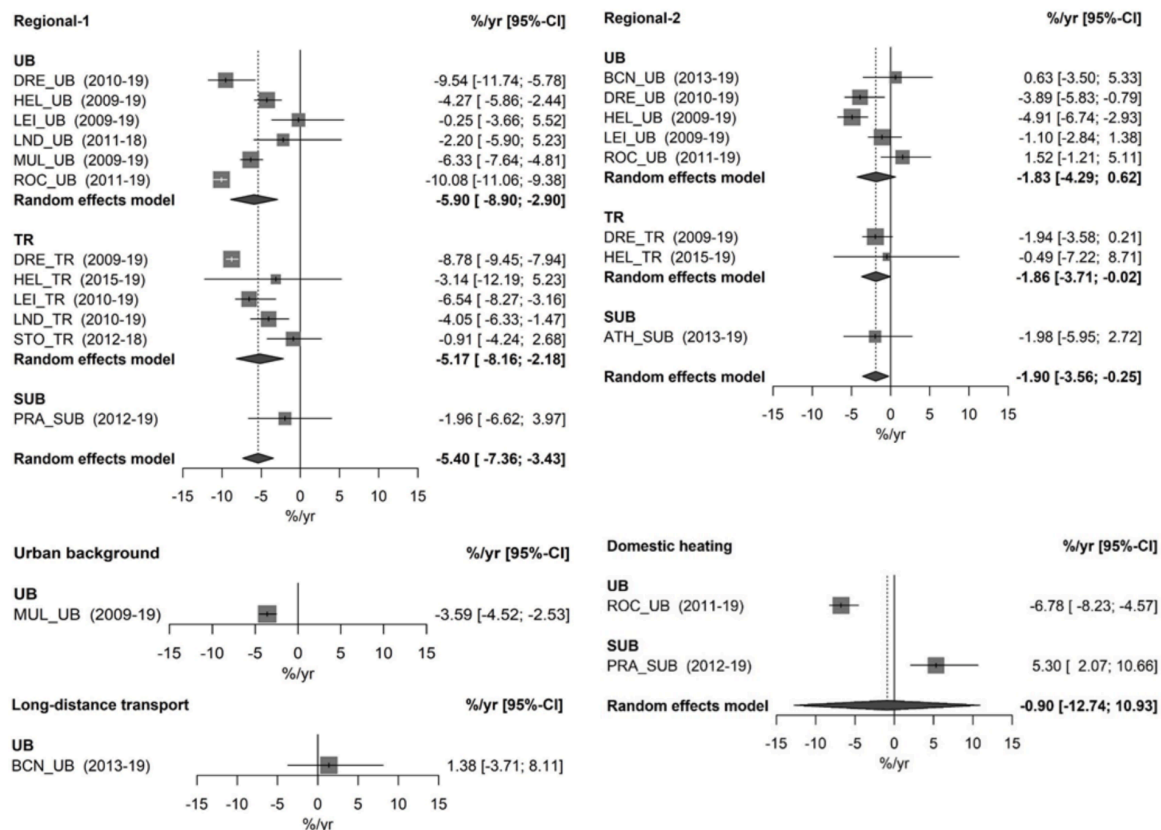


Fig. 7. Trend analysis and meta-analysis results for *Regional-1*, *Regional-2*, *Urban background*, *Domestic heating* and *Long-distance transport* sources. Trends are calculated using the Theil-Sen method. Mean effects for each site type (Urban background, UB; Traffic, TR; Suburban background, SUB; and Regional background, RB) are estimated using a random effects model. The meta-analysis results are presented for each pollutant across the different site types, as well as for global trends (dashed line).

implementation of European emission standards, especially after the introduction of diesel particle filters (DPFs) from the 2011 EURO 5/V standard for primary particles. The effect of DPFs is observed in the higher reduction of *Traffic-2* (primarily associated with diesel vehicle emissions) compared to *Traffic-1* (mainly gasoline vehicle emissions and nucleation of UFP from diesel). Furthermore, the reduction of sulphur (S) content in diesel might have contributed to reducing UFP from road traffic. However, ultra-low-S diesel (ULSD) was implemented in 2006 in Europe and the US, which was before the period covered by the datasets in this study. Other road traffic policy measures, such as Low Emission Zones (LEZ) in many cities and the Congestion Charge in some of them, also might have contributed to this reduction.

Traffic-Nucleation (and *Traffic-1* to a lesser extent) included the nucleation of particles generated during the dilution of diesel exhaust semi-volatile organic compounds (SVOCs), thus DPFs cannot retain them. This has prompted lighter decreasing trends for *Traffic-Nucleation* and *Traffic-1* ($-2.97\% \text{ yr}^{-1}$ and $-2.78\% \text{ yr}^{-1}$) as compared with *Traffic-2* ($-4.21\% \text{ yr}^{-1}$). Emission control technologies should be applied to DPFs to reduce the emissions of SVOCs. Furthermore, DPFs might be less efficient for particles smaller than 23 nm in diameter ($N_{<23}$). These results indicate that EURO standards should include emission controls for particle number concentration (PNC) for at least particles > 10 nm and, if possible, for > 1 nm, and not only for > 23 nm, as the current standards have.

Trends in *Photonucleation* were more diverse among the sites. In some cases, this source increased probably due to a reduction in condensation sink, which facilitates photochemical NPF. In other cases, the reductions might be associated with the decrease of the emission of precursors of

newly formed particles such as SO_2 . The decrease in primary PM emissions and precursors of UFP also contributed to the reduction of urban and regional background sources.

The results of the study demonstrate the positive abatement of UFP in Europe, but more measures should be applied to reduce traffic emissions in the cities, especially an effort should be made to implement policies to reduce nucleated particles, both delayed primary diesel traffic particles and those from photochemical nucleation.

CRediT authorship contribution statement

Meritxell Garcia-Marlès: Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Rosa Lara:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Cristina Reche:** Writing – review & editing, Data curation. **Noemí Pérez:** Writing – review & editing, Data curation. **Aurelio Tobías:** Writing – review & editing, Methodology. **Marjan Savadkoobi:** Writing – review & editing, Data curation. **David Beddows:** Writing – review & editing, Data curation. **Imre Salma:** Writing – review & editing, Data curation. **Máté Vörösmarty:** Writing – review & editing, Data curation. **Tamás Weidinger:** Writing – review & editing, Data curation. **Christoph Hueglin:** Writing – review & editing, Data curation. **Nikos Mihalopoulos:** Writing – review & editing, Data curation. **Georgios Grivas:** Writing – review & editing, Data curation. **Panayiotis Kalkavouras:** Writing – review & editing, Data curation. **Jakub Ondracek:** Writing – review & editing, Data curation. **Nadezda Zikova:** Writing – review & editing, Data curation. **Jarkko V. Niemi:** Writing – review &

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2024.109149>.

Data availability

Data will be made available on request.

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4.2.4 Paper IV

Spatiotemporal variation and source apportionment of ultrafine particles near Brussels Airport (Belgium)

Garcia-Marlès, M., Peters, J., Van Laer, J., Frijns, E., Berghmans, P., Staelens, J., Hofman, J., de Vos, T., Brasseur, O., Van Poppel, M., Alastuey, A., Querol, X.

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Spatiotemporal variation and source apportionment of ultrafine particles near Brussels Airport (Belgium)

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HIGHLIGHTS

- Aircraft emissions impact ultrafine particle (UFP) near Brussels Airport.
- UFP contributions decreased with distance but persisted up to 7 km from the airport.
- Airport sources contributed 6500–14000 # cm⁻³ near the airport.
- These contributions are diluted rapidly due to atmospheric dispersion.
- Takeoff emissions are generally coarser (20 nm) than taxiing (12 nm).

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ABSTRACT

Ultrafine particle (UFP) emissions from aircraft engines are associated with increased UFP concentrations near airports, raising concerns for air quality and public health. This study investigated the impact of airport operations at Brussels Airport (Belgium) on local air quality during a two-month monitoring campaign in autumn 2015. Four monitoring sites, strategically located from the airport and along a transect parallel to the main runway, were used to measure particle number size distributions (PNSD), black carbon (BC), particulate matter (PM₁₀), and nitrogen oxides (NO_x). This novel approach, using high-resolution PNSD data, provided a detailed analysis of aircraft-related UFPs under varying meteorological conditions and operational phases. Results showed strong correlations between UFP concentrations, meteorological conditions, and aircraft landing and takeoff (LTO) operations, with the smallest measured particles (10–20 nm) dominating the particle number concentrations (PNC) near the airport, contributing up to 65 % of total PNC. Aircraft emissions were responsible for 15000–20000 # cm⁻³ of 10–20 nm particles near the airport, with peak concentrations (30-min) reaching 65000–80000 # cm⁻³ during high-activity periods. While airport-related UFP concentrations decreased with distance, they persisted up to 7 km. Source apportionment analysis identified taxiing and takeoff as major contributors to UFPs near the airport (50–64 % of PNC), with road traffic, biomass burning, and regional background more significant at greater distances. Despite attempts to distinguish between landing and takeoff emissions, similar particle profiles were observed for both operations. These findings provide insight into the spatial persistence, dominant operational phases, and size distribution of aircraft-related UFP emissions, and underscore the need for targeted mitigation strategies to improve air quality near airports.

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1. Introduction

Ultrafine particles (UFPs) comprise aerosols with a diameter smaller than 100 nm. In ambient air, UFPs are typically high in numbers but low in mass (Baldauf et al., 2016), thus their concentrations are most commonly (but not exclusively) measured in terms of particle number concentration (PNC) per unit volume of air ($\# \text{ cm}^{-3}$). As a result, UFPs are often underrepresented in conventional air quality monitoring, which focuses primarily on mass-based metrics (de Jesus et al., 2019). As recognised by WHO (2021), there is a body of evidence on the health effects for UFPs but results are inconsistent. Several studies have reported that the size of UFPs allows them to reach the deeper parts of the respiratory system, translocating through the alveoli and reaching the circulatory system (Cassee et al., 2019; Kreyling et al., 2014; Oberdörster et al., 2005; Peters et al., 2006). Oberdörster et al. (2004) also showed that UFPs might directly translocate through the olfactory bulb and reach the brain. Furthermore, a number of studies found a positive relationship between exposure to UFPs and increased premature mortality (Ibald-Mulli et al., 2004; Schwarz et al., 2023; Tobías et al., 2018; Wichmann et al., 2000).

Knowledge about the number and size distribution of specific UFP sources, as well as their formation and transformation processes, has increased significantly over the past years. Most studies conducted in urban environments point to traffic as a major source of UFP exposure (Zhou et al., 2005; Pey et al., 2009; Dall'Osto et al., 2012; Harrison et al., 2011; Kumar et al., 2014; Brines et al., 2015; Beddows and Harrison, 2019; Rivas et al., 2019; Hopke et al., 2022; Kalkavouras et al., 2024; Garcia-Marlès et al., 2024). Other sources and processes, such as new particle formation (NPF), aircraft emissions, shipping, and domestic and regional sources, also contribute, with aviation identified as the third largest source of UFPs in Europe after shipping and road transport (Kuenen et al., 2022).

Although concerns about UFP emissions from aircraft have been increasing in recent years, there are few studies evaluating UFP emissions in and around airport areas. Some studies have evaluated the effects of UFPs emitted from airports in cities around the world (Cheung et al., 2011; Hofman et al., 2016; Hudda et al., 2014; Keuken et al., 2015; Masiol et al., 2017; Rivas et al., 2019; Westerdahl et al., 2008). Other studies were performed within airport grounds (Campagna et al., 2016; Hsu et al., 2013; Masiol et al., 2016; Mazaheri et al., 2009; Moore et al., 2017; Psanis et al., 2017; Stacey et al., 2020, 2023; Stafoggia et al., 2016; Yu et al., 2019) or in the surrounding areas (Dröge et al., 2024; Fushimi et al., 2019; Hsu et al., 2012; Hudda et al., 2018; Pirhadi et al., 2020; Takegawa, 2023; Tremper et al., 2022).

In a series of experiments by the US EPA, the effects of fuel consumption, fuel type, engine type, engine cycle, and engine temperature on emissions were investigated (Kinsey et al., 2010). It was observed that emissions from aircraft engines are high, in the range of 10^{15} to 10^{17} particles per kg of fuel consumed. The size distribution of these particles exhibited a single mode between 3 and 100 nm, with a geometrical median diameter of particles from 10 to 30 nm. Furthermore, aircraft fuel combustion has been linked to the emissions of particulate matter (PM), black carbon (BC), nitrogen oxides (NOx), ozone (O_3) precursors, sulphur dioxide (SO_2), and carbon monoxide (CO) (Carslaw et al., 2006; Hu et al., 2009; Lobo et al., 2012; Masiol and Harrison, 2014; Westerdahl et al., 2008). Ellermann et al. (2012) revealed that UFP concentrations were 3–4 times higher at the airport in Copenhagen than at an urban traffic location, and UFPs with diameters from 6 to 40 nm were most prevalent at the airport compared to larger sized particles, accounting for 85–90 % of the total PNC at the airport apron. Stafoggia et al. (2016) and Keuken et al. (2015) found significant UFP increases under downwind conditions at the airports of Ciampino (Rome) and Schiphol (Amsterdam), respectively. Hofman et al. (2016) estimated

Schiphol airport to contribute 5 % of the total and 16 % of the 10–20 nm sized UFP number concentrations measured in the city centre of Amsterdam, 8 km from the airport. A comparable 4- to 5-fold increase in total particle numbers was observed near Los Angeles International Airport at 8–10 km downwind of the airport (Hudda et al., 2014). Thus, studies near airports confirmed aviation to be a source of mainly smaller-sized (<40 nm) UFPs.

Accordingly, UFP concentrations are highly variable, influenced by aircraft operations, weather conditions, and proximity to airports. Identifying the sources of particle number size distributions (PNSDs) is fundamentally important since particle emissions can vary significantly depending on the aircraft operation (taxiing, takeoff, landing, idling), engine type, and fuel used. However, real-time PNSD measurements of aircraft emissions at airports are scarce and typically do not differentiate between the different phases of aircraft activity or other sources, and they often start measurements from a relatively large lower size limit of particle detection (e.g., $d_{50} > 14\text{--}15$ nm, as in Masiol et al., 2016; Stacey et al., 2020; Tremper et al., 2022), meaning particles smaller than this threshold are not efficiently detected.

The aim of this study is to assess the local spatial and temporal variation of UFPs in the vicinity of Brussels Airport based on a 2-month monitoring campaign at four sites. The measurement sites were strategically located along a transect that aligns with the airport's main runway, allowing for a detailed analysis of downwind and upwind effects under varying wind conditions. This novel design provides a unique opportunity to isolate and assess the specific impact of airport operations on ambient ultrafine particle concentrations. UFP dynamics are compared to meteorological conditions and airport activity to estimate the net contribution of aircraft operations. Ambient UFP concentrations are further evaluated against particulate matter (PM_{10}), black carbon (BC), and nitrogen oxides (NOx) to contextualize the airport's contribution relative to other dominant sources such as road traffic. The study design, which includes continuous and stationary monitoring of multiple pollutants at all four sites throughout the entire campaign period, enables robust temporal analysis and source characterization across different locations. Finally, source apportionment of PNSD is performed to identify and quantify aircraft emission processes such as taxiing and takeoff, and other sources contributing at the airport. This analysis is conducted using the receptor model Positive Matrix Factorization (PMF; Paatero and Tapper, 1994).

2. Methodology

2.1. Monitoring sites

Brussels Airport is embedded in a diverse landscape of commercial, residential, and agricultural areas, with major highways surrounding it. The area represents a multi-source environment for air pollution. The monitoring program was designed to distinguish the airport's contribution within this complex setting. Therefore, four locations (named A, B, C, and D from southwest to northeast) were selected based on their proximity and varying distances from the busy runways (25R/07L, Fig. 1). Runway 25R is predominantly used for landing and takeoff under southwest wind conditions, whereas runway 07L is predominantly used under northeast wind conditions.

The selection of the four sites along a near-linear transect provided an opportunity to examine the spatial variability of ultrafine particle concentrations in direct relation to airport operations and meteorology. This setup enabled a systematic comparison between upwind and downwind sites under different wind regimes, offering insights into dispersion patterns and the relative influence of aircraft activities. To our knowledge, this type of transect-based design, aligned with a major runway at a busy international airport, has not often been used in prior

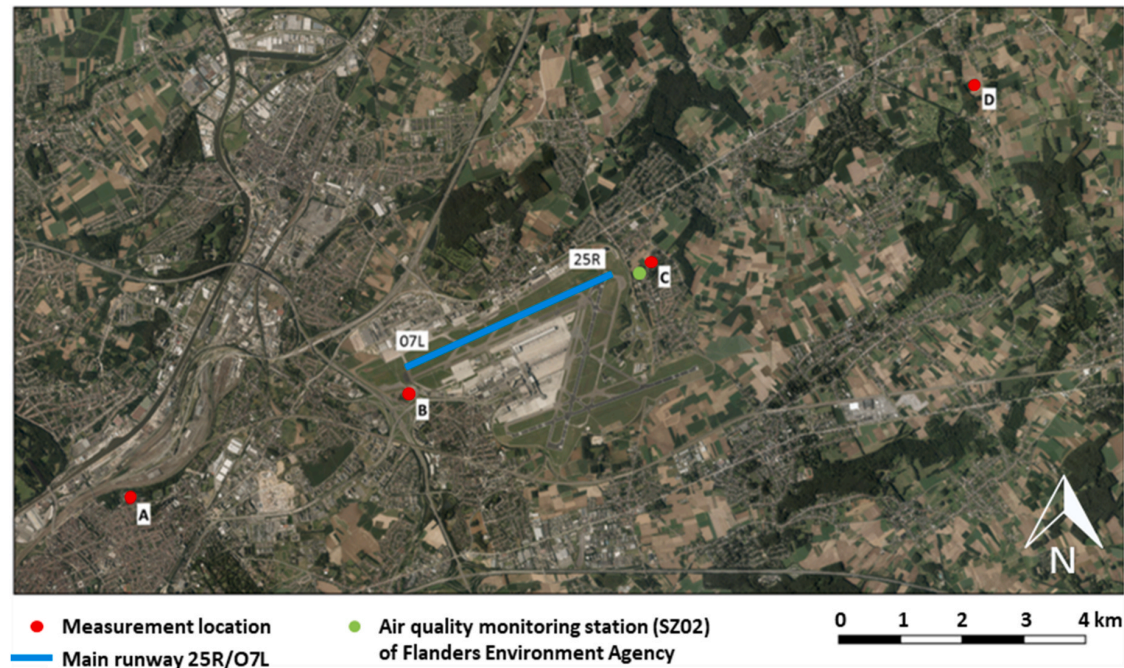


Fig. 1. Overview of the four monitoring locations (A, B, C and D) at varying distance from the main occupied runway 25R/07L, along with the SZ02 air quality monitoring station operated by the Flanders Environment Agency. Close-up maps of each site are provided in [Supplementary Fig. S1](#).

UFP monitoring studies, and adds a novel dimension to the characterization of airport emissions.

The monitoring campaign took place over a 2-month period in autumn 2015 (from October 1st to November 30th, 2015). Prior to the main campaign, collocated measurements were conducted at location C for 2 weeks to evaluate the comparability of the monitoring equipment used.

Locations A and B are situated upwind from the airport during southwest wind conditions (the most common wind direction), whereas locations C and D are downwind of the airport under these conditions. Location C is situated 150 m northeast from an existing air quality monitoring station (SZ02) of Flanders Environment Agency. In terms of land use, locations A and C can be categorized as urban background sites, and location D as a rural site. Monitoring station B is an urban area, 200 m from Brussels' main ring road, which experiences very high traffic volumes (e.g., >100000 vehicles per day, or an estimated hourly average of >4000 vehicles). During southwest wind conditions, road traffic sources are expected to dominate at site B, whereas under northeast wind conditions, the influence of airport activities is anticipated to be more significant. Photos of the monitoring sites are provided in [Supplementary Material S1](#).

2.2. Instrumentation and data

The monitoring focused on UFP, NO_x, BC and PM₁₀ (Table 1). UFP number concentrations were measured using Scanning Mobility Particle Sizer (SMPS) monitors at the four locations with a time resolution of 5 min. This high temporal resolution was chosen to capture rapid fluctuations in UFP concentrations, particularly from episodic sources like aircraft emissions, which can vary significantly over short timescales. A 5-min resolution provides a balance between detecting these short-term peaks and maintaining data reliability. Particle number counts were corrected for diffusion losses using the equations of [Gormley and Kennedy \(1949\)](#) for the IfT custom classifier and CPC3772 at sites A and B, or using equations from [Hinds \(1999\)](#) for the TSI SMPS 3936L76 at sites C and D. Particle number counts were aggregated into seven size classes:

Table 1
Overview of the instrumentation. Ultrafine particles, UFP; Southwest, SW; Northeast, NE.

Site	Variable	Size selection, tubing, flow	Monitor
A (5 km SW of airport)	UFP	PM ₁₀ inlet (2.3 m ³ h ⁻¹), isokinetic splitting (5 points), Nafion dryer	IfT custom classifier and CPC3772
	BC	No size-selective inlet	Magee AE22
	NO _x	Teflon tubing	NO _x TS42C
	PM ₁₀	PM ₁₀ inlet (2.3 m ³ h ⁻¹)	Leckel SEQ47/50, EN12341:2014
B (250 m SW of airport)	UFP	PM ₁₀ inlet (2.4 m ³ h ⁻¹), isokinetic splitting (5 points), Nafion dryer	IfT custom classifier and CPC3772
	BC	No size-selective inlet	Magee AE22
	NO _x	Teflon tubing	NO _x 42C
	PM ₁₀	PM ₁₀ inlet (2.3 m ³ h ⁻¹)	Leckel SEQ47/50, EN12341:2014
C (750 m NE of airport)	UFP	TSI Environmental Sampling System, PM ₁₀ inlet (1.0 m ³ h ⁻¹), PM ₁ cyclone and Nafion dryer	TSI SMPS 3936L76 Kr-85 185 MBq, L-DMA 3081 < 10–1000 nm, TSI CPC 3776, butanol, 1 L/min flow
	PM ₁₀	PM ₁₀ inlet (2.3 m ³ h ⁻¹)	Leckel SEQ47/50, EN12341:2014
	BC	TSP inlet (16.7 L/min)	MAAP 5012
	NO _x	Teflon manifold and tubing	NO _x TS42i
D (7 km NE of airport)	UFP	TSI Environmental Sampling System, PM ₁₀ inlet (1.0 m ³ h ⁻¹), PM ₁ cyclone and Nafion dryer	TSI SMPS 3936L76 Kr-85 185 MBq, L-DMA 3081 < 10–1000 nm, TSI CPC 3776, butanol, 1 L/min flow
	BC	conducting silicone tubing, no size-selective inlet	MAAP 5012
	NO _x	Teflon tubing	NO _x TS42C
	PM ₁₀	PM ₁₀ inlet (2.3 m ³ h ⁻¹)	Leckel SEQ47/50, EN12341:2014

10–20, 20–30, 30–50, 50–70, 70–100, 100–200, and 200–294 nm, with the latter two classes of particles with diameters outside the upper size limit of UFP (100 nm).

NOx concentrations were measured by chemiluminescence at all four locations with a time resolution of 30 min. The NOx monitors underwent calibration (zero and span) every fortnight. BC concentrations were measured using a dual-wavelength aethalometer (Magee Scientific AE22 at sites A and B) and Multiangle Absorption Photometer (Thermo Scientific MAAP 5012 at sites C and D) with a time resolution of 30 min. PM₁₀ was monitored as 24-h average mass concentrations using the gravimetric reference method (aerosol sampled on quartz fibre filter using Leckel SEQ47/50). All monitoring equipment was time-synchronized. At site C, there was a distance of 150 m between the temporary site for UFP and PM₁₀ measurements and the long-term monitoring site for BC and NOx.

UFP number concentrations were compared among the four SMPS instruments during a 2-week pre-campaign of collocated measurements at one location (C) in September 2015. The comparison was generally good, with differences equal to or less than 8 % (further detailed in [Supplementary Material S2](#)). Although size-class specific scaling factors were derived from a linear regression analysis without intercept, they were not applied to the dataset used in this study, as rescaling was not performed on the provided data. A similar methodology was used for the BC measurements, achieving very good comparability between the instruments. After rescaling, the highest difference observed between instruments was less than 3 %.

Meteorological data were provided by the Royal Meteorological Institute of Belgium (RMI) from a synoptic station in Melsbroek (near the airport), including wind speed, wind direction, air temperature, relative humidity, and precipitation. Wind data were obtained at 10 m above ground, while temperature and humidity were measured at 1.5 m above ground, all with a time resolution of 1 h. Precipitation was measured with a 6-h time resolution. Additionally, the RMI provided data on the height of the mixing layer, based on LIDAR ceilometer measurements in Ukkel, which are post-processed by a model. More details are given by [Laffineur et al. \(2013\)](#).

Log files of landings and takeoffs at Brussels Airport were provided by the airport authorities and the Department of Environment, Nature and Energy of the Flemish Government. These data include the actual date and time of landing or takeoff and the runway used.

2.3. Representativeness of the monitoring campaign

The effect of seasonality and year-round variability of the UFP, NOx, PM₁₀, and BC concentrations in the vicinity of the airport was not within the scope of this study. However, it is important to consider the representativeness of the study period. Thus, the main meteorological variables influencing air quality were compared between the study period (October–November 2015) and the same months in the preceding years, 2013 and 2014. The temperature, relative humidity, wind speed, amount of rainfall, and the number of rainy days were within the expected range ([Supplementary Material S3](#)). Wind direction was also comparable with observations from 2013. In October–November 2014, the dominant wind direction was SE, in contrast to SW for October–November 2013 and 2015. Over a whole year, SW wind directions are nevertheless the most common in the study region. Thus, generally, the air quality monitoring campaign took place under representative meteorological conditions.

One of the main hypothesized sources of UFP, aircraft emissions, was compared for the period October–November between 2015 and 2016. In October and November 2015, 19834 aircrafts took off and 19843 aircrafts landed (total 39677). These numbers of operations are comparable to October–November 2016 when 19449 aircrafts took off and 19468 aircrafts landed (total 38917).

2.4. Data analysis

The data analyses were performed in R ([R Core Team, 2016](#)). General summary statistics were used to explore the time series measurements, focusing on 95th and 99th percentile values as peak events. Boxplot analyses were employed to compare the spatial and temporal variations of parameters. Pollution roses were used to study pollutant concentrations as a function of wind direction, using the *Openair* package ([Carslaw and Ropkins, 2012](#)). Other visualizations were used to examine the combined effects of meteorology and airport activity on pollutant concentrations. Additionally, a multilinear regression analysis was conducted to identify the most significant variables explaining UFP dynamics. Finally, correlation analyses were performed between different UFP size classes and other pollutants (NOx and BC).

2.5. Positive matrix factorization

Positive Matrix Factorization (PMF; [Paatero and Tapper, 1994](#)) was applied for the identification and apportionment of sources of PNSD, using the underlying solver multilinear engine 2 (ME-2, [Paatero, 1999](#)). This is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into two matrices: factor contributions and factor profiles. These matrices require interpretation to identify source types by utilizing measured source profile data and emissions inventories.

The datasets included 5-min time resolution PNSD data combined with concentrations of BC, NO, and NO₂ to help identify the sources. The analysis was conducted by combining data from the four monitoring sites so that PMF solved factors for all the sites simultaneously. Time series for each site were concatenated, ensuring that the final dataset had the same size bin columns across all sites. As a result, the factor profiles (i.e., the PNSD) would be the same between the sites. With this approach, site-specific temporal variations could be accounted for while maintaining consistent source profiles, enabling a more robust comparison of sources across the different sites. Furthermore, the simultaneous analysis of all sites helped improve the stability of the PMF solution, as it leveraged a larger dataset, thus enhancing the identification of key contributing factors and their regional influences.

The measurement uncertainties of PNSD were calculated using the methodology established by [Ogulei et al. \(2007\)](#) and further developed by [Rivas et al. \(2019\)](#), with the following equation:

$s_{ij} = \alpha_{ij} + C_3 \cdot n_{ij}$ where $\alpha_{ij} = 0.01(n_{ij} + \bar{n}_{ij})$, n_{ij} is the concentration of sample i in size bin column j , $\bar{n}_{ij}$ is the arithmetic mean of concentration in size bin j , and C_3 is a constant determined by trial-and-error and set to 0.05 in this study. Uncertainties for the ancillary co-pollutants were calculated by [Polissar et al. \(1998\)](#) equation. An additional uncertainty of 0.07 was applied to all variables to optimise the model.

PMF was run multiple times, and the optimal number of sources was identified through trial and error by adjusting the number of factors. A six-factor solution was chosen based on accepted criteria and guidelines ([Belis et al., 2019](#); [Hopke et al., 2023](#)), considering a $Q_{\text{true}}/Q_{\text{exp}}$ ratio close to 1 (0.98 in this study), profile uncertainties evaluated using the displacement method, and the ability to provide the most meaningful physical interpretation with reasonable residuals. The contributions of each factor to the variance of the ancillary co-pollutants, along with daily patterns, seasonality, and polar plots, were assessed to further support the identification of the sources.

3. Results and discussion

3.1. Overview of UFP number concentrations at the different locations

To allow for comparison, only measurements taken simultaneously at all four locations were included in this analysis (17278 5-min measurements over nearly 60 days). The average total PNC (from 10 to 294 nm) was lowest at the rural site D (7700 # cm⁻³) ([Fig. 2](#)).

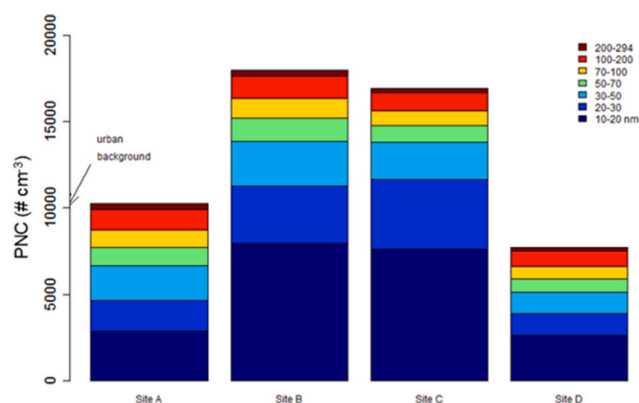


Fig. 2. Average particle number concentration (PNC) of the different size classes at the four monitoring locations, based on 5-min measurements over a 2-month period. An estimation of the urban background concentration of 10 000 # cm⁻³ is given for comparison.

Concentrations were significantly higher at the other sites, especially near the airport. At sites C and B, the average concentration was 2.2 and 2.4 times higher, respectively, compared to the rural site D, while at urban site A, it was 1.3 times higher. Peak concentrations, given as 99th percentile concentrations (P99), were 3.7 and 4.2 times higher at sites B and C compared to site D, while at site A (urban) peak concentrations were 0.7 times lower than at site D (more influenced by the airport).

The urban background concentration of 10 000 # cm⁻³, shown in Fig. 2, was estimated as a reference value, representing a long-term average urban background concentration for PNC, derived from literature values for European cities (Hofman et al., 2016; Trechera et al., 2023). It provides a reference for comparison, reflecting typical urban background levels without significant local sources like airports. However, it is important to note that urban background concentrations are not static and can vary significantly due to diurnal patterns, meteorological conditions and other factors. The fixed value of 10 000 # cm⁻³ should therefore be interpreted as a simplified estimate. At Site A, identified as the urban site, the average PNC (approximately 10 000 # cm⁻³, as derived from Fig. 2) aligns closely with this estimate. In contrast, rural site D showed a lower average concentration (4000 # cm⁻³), consistent with rural background PNC levels reported in other European regions. For example, average rural background PNCs in Germany ranged between 3000 and 5000 # cm⁻³ (Birmili et al., 2016), while a rural site in Midland Switzerland reported an average of 4000 # cm⁻³ (Dada et al., 2025). These findings suggest that, although site D is occasionally influenced by emissions from the nearby airport, its average PNC remains within the typical rural range, likely because the airport's impact is short-lived and sporadic. However, to better assess this influence, longer measurement periods would be necessary.

For the smallest particles (10–20 nm), frequently associated with aircraft emissions (Keuken et al., 2015; Masiol et al., 2017; Ridolfo et al., 2024; Stacey, 2019; Tremper et al., 2022), average and peak

concentrations are detailed in Table 2. Near the airport (sites B and C), average concentrations of the 10–20 nm size class were around 8000 # cm⁻³, three times higher than at the rural site D (around 2800 # cm⁻³), and comparable to the urban site A. Peak 5-min concentrations at sites B and C reached 69 000 and 74 000 # cm⁻³, respectively, which is over three times higher than at site D (20 000 # cm⁻³). However, peak concentrations at site D were still nearly twice those observed at the urban site A (10 000 # cm⁻³), suggesting that despite its rural classification, site D was not at urban background levels and was influenced by episodic UFP emissions from the airport. The ratio of peak to average concentrations further illustrates variability: 8.4–9.5 at sites B and C, 7.5 at site D, and 3.5 at site A. This indicates a large variability in UFP concentrations near the airport (sites B and C) and to a lesser extent at the rural site D, with lower variability at the urban site A.

The proportion of UFPs in different size classes varied across the four locations (Fig. 2). The 10–20 nm size class dominated near the airport (sites B and C), representing 45 % of the total 10–294 nm particles. At site D and A, this size class represented 35 % and 28 %, respectively. The highest concentration of particles larger than 30 nm was at site B, followed by sites A and C.

UFP concentrations showed a bimodal daily pattern (Fig. 3 and Supplementary Material S4), peaking between 6 and 10 a.m. and 4–8 pm. This pattern was consistent across all UFP size classes, including 10–20 nm particles. A similar bimodal daily pattern is observed in PM and UFP time series from road traffic sites, due to the increased traffic intensity during morning and evening rush hours (Trechera et al., 2023) and the thicker planetary boundary layer at midday (diluting UFP). Aircraft takeoffs and landings also followed this bimodal pattern at all sites (Supplementary Material S4), making it difficult to distinguish between road traffic and aircraft sources. At the rural site D, UFP concentrations showed less pronounced variability during daytime hours compared to the other sites, with no clear peaks corresponding to rush hours (Supplementary Material S4).

3.2. Overview of BC, PM₁₀ and NO_x concentrations at the different locations

In contrast to UFP, BC mass concentrations (with a major road traffic origin) were similar across A, B, and C locations (Table 3). At the rural site D, BC concentrations were significantly lower, with 30-min peak levels being 2 to 3 times lower than those observed at the urban sites. Despite these differences in absolute values, day-to-day variations in BC concentrations were consistent across all sites. Sites B and C exhibited similar daily patterns, with higher concentrations during traffic rush hours (Supplementary Material S5), likely due to emissions from (relatively old) diesel vehicles. Diesel exhaust produces BC-rich soot particles, especially in the absence of diesel particle filters (DPFs) (Cédric et al., 2016). These emissions arise from vehicles associated with airport operations and road traffic on the surrounding roads. Site A experienced consistently high BC concentrations throughout the day, while site D showed elevated levels during traffic rush hours and at night. The night-time increase at site D may be attributed to domestic heating emissions, which release BC as a result of incomplete combustion of carbon-based fuels such as wood. While BC is primarily attributed to road traffic and residential heating at the study sites, it is known that aircraft can also emit BC (Hudda et al., 2020; Stacey et al., 2021). However, in the presence of multiple combustion sources, clear attribution of ambient BC to aircraft activity is difficult (Tremper et al., 2022).

Daily average PM₁₀ concentrations varied regionally (Table 3). Amato et al. (2016) reported that secondary PM₁₀ and PM_{2.5} were dominant contributors to annual averages in 5 cities, and these secondary PM concentrations also exhibited regional-scale variability. The lowest PM₁₀ concentrations were observed at the rural site D and near the airport (site C), suggesting that airport activities did not contribute significantly to increased PM₁₀ levels.

Table 2

Average and P99 (peak concentrations, given as 99th percentile) concentrations particle number concentration (PNC) of the 10–20 nm size class at the four monitoring locations. The ratios in comparison to the location with the lowest average concentration (rural site D) are also given.

Site	Average of 10–20 nm		P99 of 10–20 nm	
	PNC (# cm ⁻³)	ratio	PNC (# cm ⁻³)	ratio
A	2900	1.1	10000	0.5
B	8100	3.1	69000	3.5
C	7800	3.0	74000	3.7
D	2600	1.0	20000	1.0

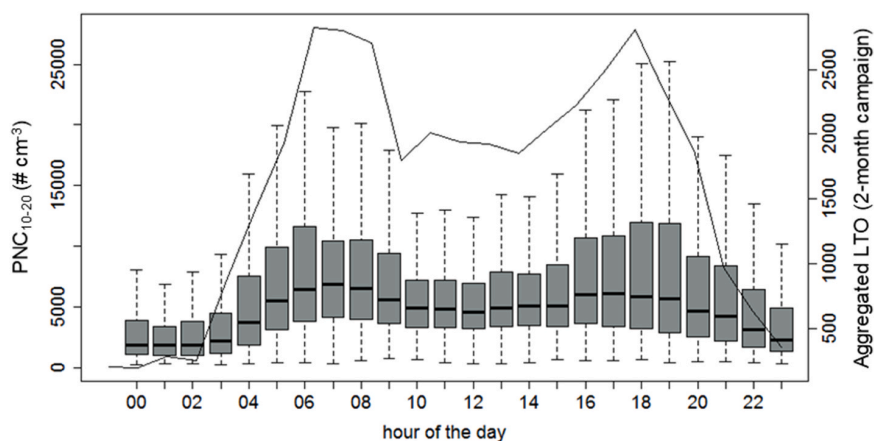


Fig. 3. Boxplot representation of the 10–20 nm particle number concentration (PNC) for each hour of the day at location B near the airport. The solid line on the secondary y-axis represents the total number of landing and takeoff (LTO) operations per hour, aggregated over the two-month measurement campaign. Similar plots for the other three locations A, C and D are given in [Supplementary Material S4](#).

Table 3

Summary statistics of the BC, NO_x and PM₁₀ concentration ($\mu\text{g m}^{-3}$) at the four locations based on 30-min averaged measurements.

Variable	Statistic	Site A	Site B	Site C	Site D
BC	1st quartile	0.9	0.9	0.8	0.5
	Median	1.4	1.4	1.3	1.0
	Mean	1.9	1.9	1.8	1.3
	3rd quartile	2.4	2.4	2.3	1.8
	Maximum	17	17	13	6.7
NO _x	1st quartile	27	33	19	12
	Median	45	56	32	22
	Mean	65	83	48	30
	3rd quartile	76	98	56	35
	Maximum	830	1400	490	340
PM ₁₀	1st quartile	17	16	15	14
	Median	21	21	19	18
	Mean	24	25	21	19
	3rd quartile	30	31	25	24
	Maximum	48	69	40	40

High NO_x concentrations were recorded at site B, located near the airport, with an average of $83 \mu\text{g m}^{-3}$, compared to $30 \mu\text{g m}^{-3}$ at the rural site D ([Table 3](#)). This is likely because site B was situated approximately 200 m from the traffic-intensive Brussels ring road (R0). Elevated NO_x levels were also observed at sites A and C (65 and $48 \mu\text{g m}^{-3}$, respectively), indicating that NO_x contributions from aircraft emissions were relatively limited compared to those from road traffic. Day-to-day NO_x concentration variations were similar across sites A, B, and C. As expected, NO_x levels at all sites displayed bimodal patterns typical of traffic rush hours ([Supplementary Material S6](#)), consistent with road traffic contributions ([de Foy, 2018](#)).

3.3. Analysis of the UFP concentration in function of meteorology and airport activity

3.3.1. UFP concentration as a function of meteorology

Meteorological data were linked to UFP measurements by time, with UFP data at 5-min intervals and meteorology at 1-h intervals. For the 10–20 nm size class, the highest PNCs occurred when the site was downwind of the airport ([Fig. 4](#)): sites C and D under SW wind, and sites A and B under NE wind. This downwind effect was observed both near

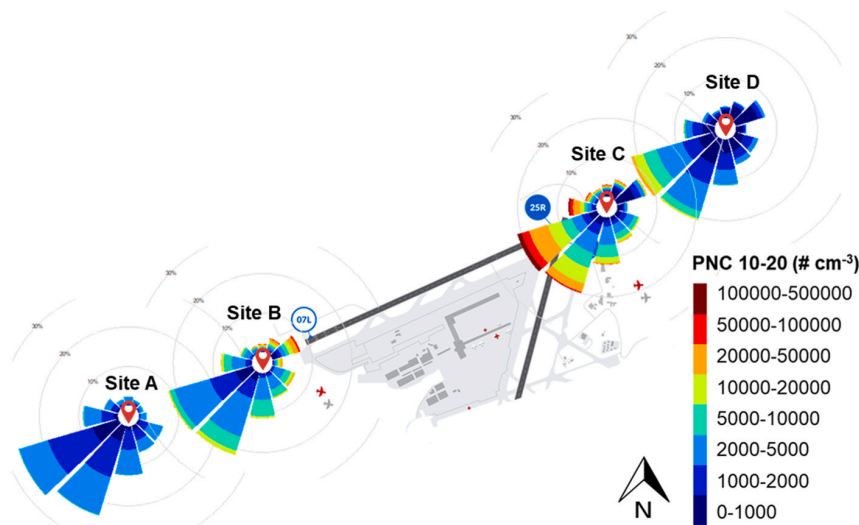


Fig. 4. Pollution roses of 10–20 nm PNC at the four sites along a SW - NE transect. The pollution roses represent the frequency distribution of wind directions. For each of the segments the colouring is proportional to the time distribution of the concentrations under downwind conditions (in $\# \text{ cm}^{-3}$, 5-min time resolution).

and far from the airport, though PNCs were much lower at the more distant sites. Under downwind conditions, peak 5-min concentrations over 50000 # cm⁻³ were found for 11 % and 6 % of the time at the sites B and C, respectively. Sites A and D never exceeded 50000 # cm⁻³, but at the rural site D, 7 km from the airport, peak 5-min concentrations of 20000 # cm⁻³ were only observed under airport downwind conditions. PNCs decreased with distance from the airport, although site D often had higher concentrations than site A, which was closer to the airport.

Similar downwind increases were observed for the 20–30 nm size class, with lower concentrations. For larger particles (>70 nm), the wind direction effect was less significant, indicating the contribution from multiple sources. Peak concentrations for these larger particles occurred in various wind directions.

The monitoring sites' alignment with the main runway and wind directions allowed distinguishing airport contributions from other traffic sources using an additive model. This model assumes that UFP concentrations at a given location results from the addition of all the upwind sources. Upwind sources other than the airport were measured at upwind sites near the airport (site B for SW wind and site C for NE wind). Subtracting the upwind concentrations from the downwind concentrations results in an estimation of the airport's net contribution. However, this simplification may underestimate airport impacts because it assumes an equal contribution of upwind sources other than the airport at the upwind and downwind sites, whereas some particles may grow, react, and deposit before reaching the downwind site. Additionally, the model assumes negligible airport activities at upwind sites, although there may be contributions from aircrafts taking off and flying at low altitude nearby the upwind site. This can lead to underestimating the airport's impact, especially at more distant sites.

The average net airport contribution to 10–20 nm UFP was estimated as 15000–20000 # cm⁻³ at the sites near the airport (Table 4, sites B and C). This equals to about 65 % of the total PNC (10–294 nm), compared to 45 % at the more distant sites A and D (Fig. 2). Peak contributions (P90, i.e., 10 % of the time) reached 45000–58000 # cm⁻³ at sites B and C. For distant sites, the model overcompensated and therefore only peak concentrations were calculated, estimated at 1500–5000 # cm⁻³ for 10 % of the time (P90) and 4000–9000 # cm⁻³ for 5 % of the time (P95). These findings show that the airport is a major source of 10–20 nm particles, resulting in largely increased UFP concentrations in the surrounding area.

These results are consistent with other studies. Ellermann et al. (2012) measured a 10000 # cm⁻³ contribution of mainly <40 nm sized particles at Copenhagen airport (1-h average). At 250–500 m from Santa Monica airport, concentrations of 15000–19000 # cm⁻³ were recorded (Hsu et al., 2012) (1-min time resolution), and 15000 # cm⁻³ at Schiphol airport, with 3000 # cm⁻³ at 15 km downwind (Bezemer et al., 2015) (1-h average). Much higher particle number concentrations (from 31000 to 1400000 # cm⁻³) were previously observed on airport apron and runways in Stockholm, Copenhagen, Los Angeles, Schiphol and Heathrow (ACI Europe, 2012; Ellermann et al., 2012; Möller et al., 2014; Westerdahl et al., 2008; Zhu et al., 2011) (from instantaneous peak values to hourly averages). At 8 km from Schiphol, Hofman et al. (2016) observed a 34 % and 2-fold increase in total and 10–20 nm PNCs,

respectively, while Keuken et al. (2015) reported a 3-fold increase in total PNCs at 7 km from Schiphol. Hudda et al. (2014) found a 4- to 5-fold increase in total PNCs 8–10 km downwind of Los Angeles airport. Urban roadside locations in major European cities typically reported total UFP concentrations around 30000 # cm⁻³ (Kumar et al., 2014) (1-h average), while a 2-year study at an urban background station in Antwerp found average total and 10–20 nm PNCs of 14000 and 2400 # cm⁻³ respectively (Hofman et al., 2016) (30-min average).

It is important to note that topography also plays a key role in particle dispersion. While Brussels airport is surrounded by relatively flat terrain, this is not the case for all airports. For example, at Zurich airport, which is located in a mountainous region, Zhang et al. (2020) found that despite efficient dilution and coagulation processes, airport-related contributions to total PNC remained substantial (10000–100000 # cm⁻³) at 2–4 km from the airport and were still significant up to 8 km. This highlights that local topography can affect both the extent and persistence of UFP plumes, and may influence the range at which elevated concentrations can be attributed to airport activity.

3.3.2. UFP concentration as a function of airport activity

To examine UFP concentrations as a function of airport activity, hourly data of LTO operations during the study period were used. These did not include helicopter flights and non-commercial flights, which are minor operations at the airport. The hourly meteorological data were merged with the airport activity data, and the UFP measurements were aggregated to hourly averages. Analysis of the data showed an association between wind direction, airport activity, and UFP concentrations at nearby sites (Fig. 5). At site C, PNCs were proportional to the number of LTO operations and highly influenced by wind direction, with the highest concentrations under downwind conditions. The gradient at site B was similar but less pronounced, likely due to fewer observations during downwind conditions and less frequent use of runway 07L, where site B was located near the aircraft starting point for takeoff.

Additionally, we analysed high-resolution PNSD measurements from the two closest monitoring sites (B and C) situated southwest and northeast of the airport, respectively. We first filtered aircraft landing and takeoff events from the LTO (landing and takeoff) dataset by identifying 5-min intervals with at least 70 % of operations being arrivals or departures. Wind direction was used to associate operations with each site: site B was linked to wind directions between 55° and 75°, and site C to wind directions between 235° and 275°. These intervals were then used to extract corresponding PNSD measurements. The results show that, with the approach followed, the PNSD profiles for landing and takeoff are largely indistinguishable, or that both activities result in similar distributions (Fig. S7). Given the moderate distance of the monitoring sites and the frequent overlap of landing and takeoff events within the same 5-min intervals for which PNSD data was available, we cannot exclude the possibility that the observed similarity in PNSD profiles results from dispersion effects or simultaneous operations, rather than a true similarity in emissions.

3.3.3. Important variables explaining the UFP variability

A modelling analysis was conducted to identify the most important variables explaining the UFP dynamics near Brussels airport. A linear regression model was fitted to the hourly averaged UFP concentrations (dependent variable) using the number of takeoff operations, the number of landing operations, wind direction, wind speed and the height of the atmospheric mixing layer as independent variables. Only LTO operations of the runway in line with the monitoring stations were accounted for. The modelling equation is:

$Y = b_0 + b_1DW + b_2AW + b_3S + b_4H$ where Y is the UFP concentration, D the number of departures, A the number of arrivals, W the wind direction, S the wind speed, H the height of the mixing layer, and b_0 to b_4 the modelling coefficients.

The model was first applied to the sites C and D. Both sites were in line and downwind of the main 25R runway, i.e., emissions from

Table 4

Estimated downwind contribution of the airport to the particle number concentration (PNC, # cm⁻³) of 10–20 nm particles at the four sites. NE, Northeast; SW, Southwest; *, Not estimated due to overcompensation (see text for explanation).

Wind direction	NE (55–75°) Site A	NE (55–75°) Site B	SW (235–275°) Site C	SW (235–275°) Site D
Mean	*	21000	15000	*
3rd quartile	*	28000	20000	*
P90	1500	58000	45000	5200
P95	4300	82000	66000	9400
Maximum	19000	280000	260000	37000

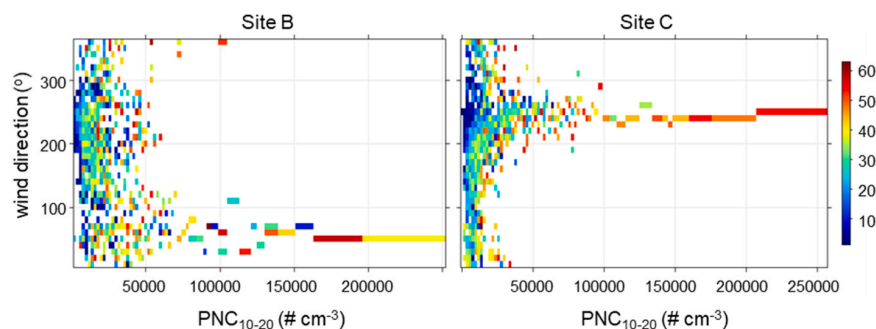


Fig. 5. Visualization of the relationship between the hourly averaged PNC of 10–20 nm particles (x-axis), the wind direction (y-axis) and the number of landing and takeoff (LTO) operations at the airport (colour scale). Site B (left) is downwind for NE wind directions (55–75°) and site C (right) is downwind for SW wind (235–275°). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

aircrafts during departure were directed towards these sites, and landing aircrafts passed at relatively low altitude over these sites. Secondly, the model was applied to sites A and B. These sites were upwind when runway 25R was used, i.e., aircrafts might have flown over these sites but could also have turned off before reaching them.

The model explained 60 % (adjusted $R^2 = 0.60$) of the variation in 10–20 nm PNC at site C. The most significant variables were the number of landings and takeoffs under SW wind direction (i.e., when site C was downwind of the airport). Wind speed, height of the mixing layer, and the number of departures or arrivals under other wind conditions were not significant. A similar model was fitted for the 10–20 nm size class at the rural site 7 km NE of the airport. This model explained a significant portion of the UFP variation (adjusted $R^2 = 0.51$). The number of LTO operations under SW wind conditions, i.e., when the monitoring site was downwind of the airport, were also significant variables. These results confirm a correlation between LTO operations at the airport and the resulting PNCs observed in the surrounding area up to 7 km from the airport.

For sites A and B (more affected by road traffic UFP), the model explained only 9 % and 32 % of the variation, respectively. The number of takeoff and landing aircrafts under downwind (i.e., SW winds) conditions were significant variables, as were wind speed and the height of the mixing layer at site B. The lower explanatory power of these models may be due to aircraft emissions being blown in the opposite direction of these sites. Additionally, the altitude of the aircraft was higher, and many aircrafts turned off before actually passing over site A. Furthermore, road traffic (not included in the model) might be a relatively more important source of UFP in these sites compared with C and D.

3.4. Correlation between UFP and other pollutants

The correlation between UFP and BC concentrations increased for

larger size classes (Table 5). At the urban background and traffic locations A and B, the correlations were low, but higher ($r = 0.22$ and 0.27 , respectively) for the smaller size classes compared to sites C and D ($r = -0.03$ and -0.21 , respectively). The positive correlation at sites A and B suggests that emissions from road traffic contributed to both BC and UFP concentrations. For the other two locations, which were mostly situated downwind of the airport (sites C and D), no or negative correlations were obtained. These results suggest a significant contribution of 10–20 nm particles originating from aircraft emissions rather than road traffic emissions. A similar conclusion could be drawn based on the correlation between UFP and NO_x, although the correlation was higher at site C.

A similar analysis was conducted for data obtained at four locations in Antwerp (VMM, 2014). The correlation between BC and the larger particle size classes was high and comparable ($r = 0.7$ to 0.9) at the sites in Antwerp and the sites of this study. For the smaller UFP size classes,

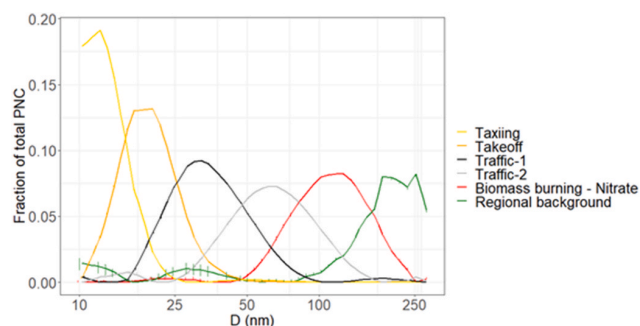


Fig. 6. Particle number size distribution (PNSD) profiles of the six sources identified.

Table 5

Pearson correlation between BC, NO_x and different particle size classes. The data for Antwerp are based on VMM (2014).

Site	Variable	Particle size class (nm)						
		10–20	20–30	30–50	50–70	70–100	100–200	200–294
Site A	BC	0.22	0.52	0.62	0.66	0.68	0.68	0.64
	NO _x	0.33	0.6	0.77	0.86	0.86	0.81	0.68
Site B	BC	0.27	0.39	0.52	0.81	0.90	0.90	0.81
	NO _x	0.26	0.36	0.51	0.77	0.81	0.76	0.63
Site C	BC	−0.03	0.05	0.39	0.75	0.84	0.87	0.76
	NO _x	0.17	0.25	0.57	0.76	0.71	0.62	0.46
Site D	BC	−0.21	0.05	0.45	0.72	0.84	0.88	0.81
	NO _x	−0.02	0.29	0.64	0.73	0.69	0.64	0.56
Antwerp		<20	20–30	30–50	50–70	70–100	100–200	>200
Urban background	BC	0.69	0.77	0.82	0.89	0.92	0.93	0.81
Park	BC	0.28	0.51	0.68	0.79	0.85	0.84	0.77
Suburban	BC	0.27	0.43	0.55	0.65	0.69	0.76	0.74
Ring road	BC	0.72	0.68	0.70	0.81	0.84	0.86	0.79

the correlations found in this study were lower than those for Antwerp, most likely because the contribution of UFP from road traffic was higher in Antwerp. The correlations for sites A and B were comparable to the values obtained at the suburban and park sites in Antwerp.

3.5. Source apportionment

Six factors (sources) of PNC were identified in the four locations of Brussels airport: *Taxiing*, *Takeoff*, *Traffic-1*, *Traffic-2*, *Biomass burning - Nitrate*, and *Regional background*. This 6-factor solution had the most physically meaningful source profiles and temporal behaviour when compared to solutions with higher or lower number of factors. The sources identified are characterised by different PNSDs (Fig. 6), daily patterns and relative contributions (Fig. 7) and polar plots (Fig. 8), with provided pollution rose plots in Fig. S8. The contribution to the variance of BC, NO, and NO₂ can be found in the supplementary material (Table S8).

3.5.1. Aircraft sources

Two UFP factors associated with aircraft emissions were identified at the sites in this study: *Taxiing* and *Takeoff*. *Taxiing* exhibited a size mode diameter of 12 nm, the finest among all the identified factors, with negligible contributions in the Aitken and Accumulation modes, while *Takeoff* exhibited a size mode diameter of 20 nm (Fig. 6). Most of the literature reports aircraft emissions as major contributors to particles <20 nm in size (Kinsey et al., 2010; Lobo et al., 2015; Masiol et al., 2016; Mazaheri et al., 2013; Ridolfo et al., 2024; Tremper et al., 2022). Additionally, several studies suggest that takeoff emissions may be coarser than the taxiing ones (Masiol et al., 2016; Ridolfo et al., 2024), likely due to differences in engine power settings and combustion efficiency. During taxiing, aircraft operate at low thrust levels (ICAO, 2022); and at these low power settings, engines exhibit reduced combustion efficiency, resulting in increased hydrocarbon emissions (Abegglen et al., 2015; Anderson et al., 2006; Brem et al., 2015). These

hydrocarbons can undergo condensation and nucleation, forming UFP in the nucleation mode (Lee et al., 2010). Numerous studies have highlighted that low thrust settings are associated with finer particle sizes, while higher thrust settings, such as during takeoff, produce coarser particles due to enhanced combustion efficiency and soot formation (Beyersdorf et al., 2014; Delhay et al., 2017; Herndon et al., 2008; Lobo et al., 2015; Vander Wal et al., 2014; Yu et al., 2017). However, both factors identified may also include particles from other sources and particles emitted during idle and landing operations, which are reported to emit lower PNC (Keuken et al., 2015; Mazaheri et al., 2009; Westerdahl et al., 2008).

Daily patterns at each site show peak contributions in the morning and evening for both aircraft factors (Fig. 7). At sites A and D (located farther from the airport), these daily patterns are less pronounced, with additional high concentrations observed at midday, which may be associated with photochemical NPF events, driven by strong solar radiation (Brines et al., 2015; Garcia-Marlès et al., 2024; Kulmala and Kerminen, 2008). *Taxiing* accounted for 23–26 % of the total PNC at sites B and C (the closest to the airport) and 14–16 % at sites A and D, while *Takeoff* contributed 27–38 % and 14–18 %, respectively (Fig. 7). Higher contributions of UFP from *Takeoff* compared to *Taxiing* have also been observed in the literature (Keuken et al., 2015; Masiol et al., 2016; Mazaheri et al., 2011). Overall, airport sources were responsible for 50–64 % of the total PNC at the sites closest to the airport and 28–34 % at the more distant sites, corresponding, in terms of PNC, to 6500–14000 and 2300–2600 # cm⁻³, respectively. These findings align with previous source apportionment studies conducted in airport environments (Pirhadi et al., 2020; Ridolfo et al., 2024; Stacey et al., 2020). However, variations between airports may occur due to differences in measurement techniques, monitoring locations, proximity to runways and taxiways, aircraft activity levels, meteorological conditions, and other emissions sources. Notably, these two factors (related to airport emissions) did not show relative contributions to any of the measured ancillary pollutants (Table S8).

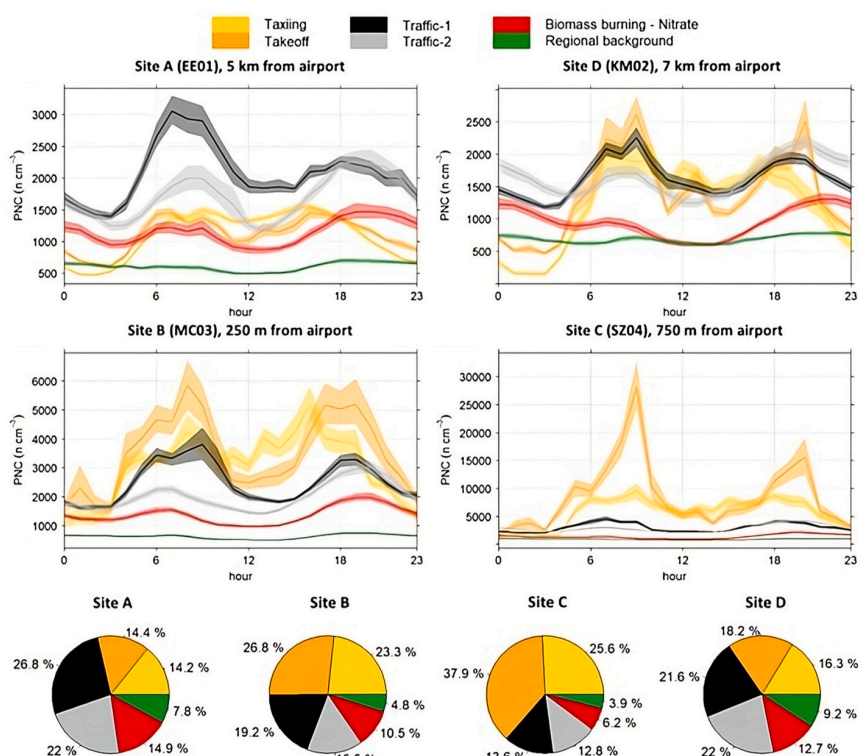


Fig. 7. Daily patterns (top) and relative contributions (bottom) of particle number concentration (PNC) of the six sources identified at each location of the study.

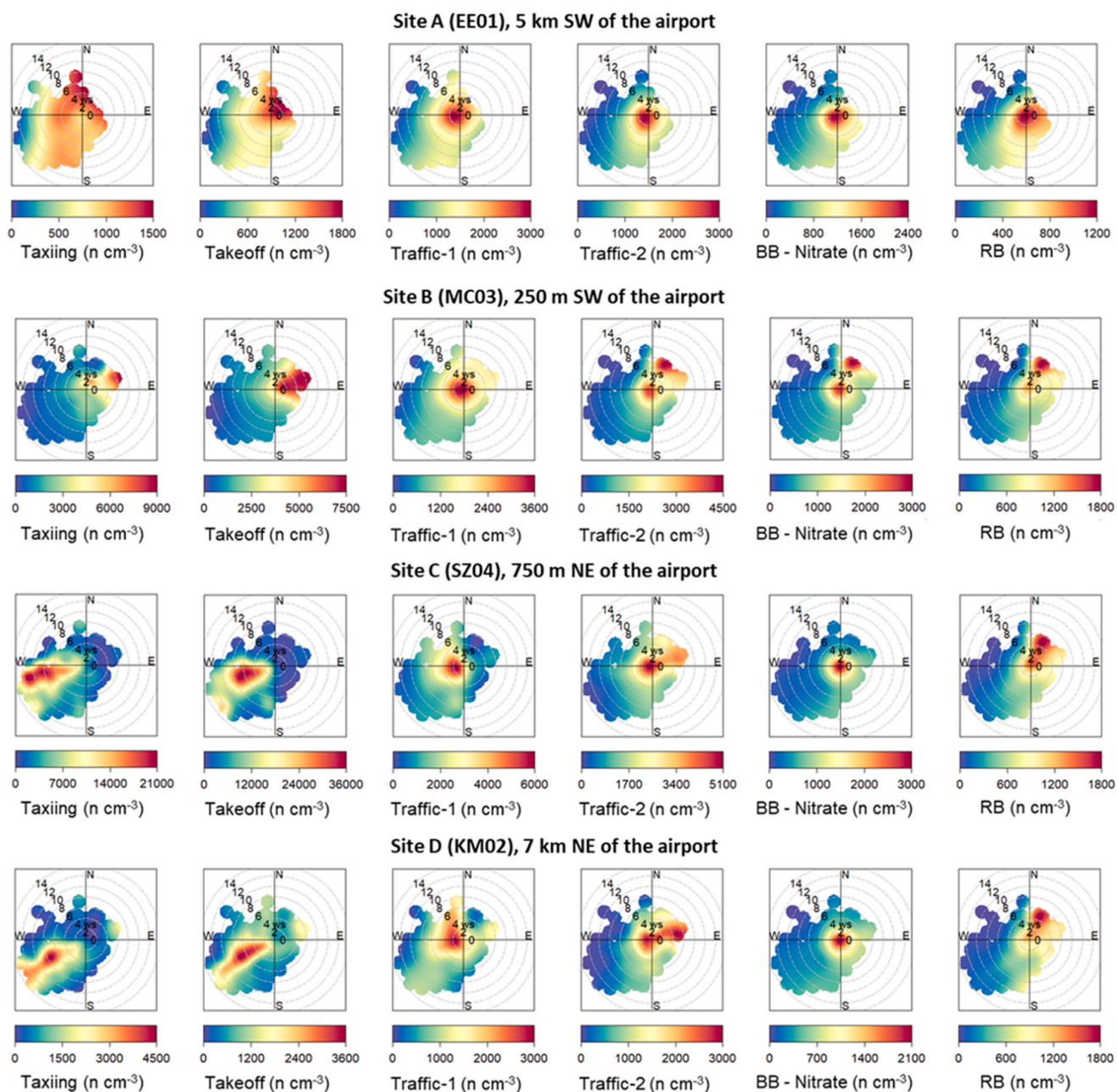


Fig. 8. Polar plots showing how contributions of the six sources identified are affected by wind direction and wind speed (m s^{-1}) at each location of the study. BB, Biomass burning; RB, Regional background.

Polar plots show similar results for both factors (Fig. 8 and S9). Sites A and B, located southwest (SW) of the airport, exhibited higher concentrations during northeastern wind conditions, coming from the direction of the airport. Conversely, sites C and D, located northeast (NE) of the airport, showed higher concentrations during southwestern winds. Thus, the airport origin is clearly evident.

3.5.2. Road traffic sources

Two traffic factors were identified, each with distinct PNSDs. *Traffic-1* is characterised by a size mode peak at 32 nm (Fig. 6) and is associated with gasoline vehicle emissions (Hopke et al., 2022; Liu et al., 2014; Ogulei et al., 2007; Vörösmarty et al., 2024) or the nucleation of particles generated during the dilution of diesel exhaust semi-volatile

organic compounds (SVOCs) (Damayanti et al., 2023; Harrison et al., 2011). *Traffic-2* is characterised by a size mode peak at 62 nm (Fig. 6) and is linked to primary diesel vehicle emissions (Hopke et al., 2022; Liu et al., 2014; Ogulei et al., 2007) or aged traffic emissions, where particles grow through coagulation as they disperse the source (Gu et al., 2011; Zhu et al., 2002).

The traffic origin of both factors is supported by the peaks observed during morning and evening rush hours (Fig. 7) and their contributions to the variance of NO_x (Table S5). Both factors contributed similarly across all sites. The combined traffic contributions accounted for 26–35 % of the total PNC at C and B (the closest to the airport) and 44–49 % at sites D and A (Fig. 7). Previous studies have reported similar traffic contributions in airport environments, varying based on the monitoring

location (Masiol et al., 2016, 2017; Pirhadi et al., 2020; Tremper et al., 2022). In contrast to airport sources, the relative contributions of road traffic sources were higher at sites further from the airport. However, sites closer to the airport showed higher absolute contributions in terms of PNC, ranging from 4500 to 5900 # cm⁻³ at sites B and C, compared to 2800 to 3300 # cm⁻³ at sites A and D. This is because of the high traffic flow associated with airport activities, including both passenger transport by road and on-site mobile machinery for aircraft ground operations, as well as the presence of large roads close to the B and C sites.

Polar plots primarily indicate a local origin, with relatively high concentrations at low wind speeds (Fig. 8 and S9), consistent with emissions from nearby roads and motorways. However, Traffic-2 also exhibited relevant contributions under NE winds, suggesting emissions from road traffic on the N21 and N26 national roads. These roads, located NE of the airport, experience daily traffic jams. In addition, numerous commercial sites along these routes also contribute to heavy traffic emissions.

3.5.3. Other sources

Biomass burning – Nitrate exhibited a size mode diameter of 126 nm (Fig. 6) and showed higher contributions at night (Fig. 7), coinciding with elevated BC levels (Table S5). This factor likely represents a mixture of sources with similar temporal and chemical characteristics rather than a single source. Specifically, it combines features typical of biomass burning associated with domestic emissions (de la Sota et al., 2018; Kuye and Kumar, 2023; Tiwari et al., 2014; Wang et al., 2020; Zhang et al., 2014), with characteristics consistent with secondary nitrate formation. Biomass burning emissions are typically higher at night (Corsini et al., 2019; Gu et al., 2011; Kleeman et al., 2009) and include BC as a product of incomplete combustion of carbon-containing fuels, such as wood and fuel oil (Chen et al., 2015; Wei et al., 2012). The presence of nitrate in this factor may be linked to secondary formation of semi-volatile ammonium nitrate, which can condense onto locally emitted particles during periods of lower temperature and higher relative humidity, which are common at night (Dall'Osto et al., 2009; Seinfeld and Pandis, 2016; Masiol et al., 2016). While the factor does not necessarily indicate a direct chemical link between biomass burning and nitrate formation, it may reflect their co-occurrence during domestic heating periods and their shared temporal and meteorological patterns. Additionally, contributions of organic nitrates or heterogeneous nitrate formation cannot be excluded, but ammonium nitrate condensation is expected to be the dominant mechanism given the observed temporal and meteorological patterns. Overall, this factor accounted for 6–15 % of the total PNC (Fig. 7), ranging between 960 and 1400 # cm⁻³, with higher contributions observed at sites A and D. Polar plots suggest a predominantly local origin, with additional contributions observed under northeastern wind conditions at site B (Fig. 8 and S9).

The final source identified, *Regional background*, exhibited a size mode diameter of 252 nm (Fig. 6) and showed slightly higher contributions also during night-time (Fig. 7). This source likely includes regional nitrates, which typically consist of particles in the Accumulation mode formed through the transport and chemical reactions of precursor gases, such as NH₃ and NO_x (Kasumba et al., 2009; Squizzato et al., 2019). This source accounted for 4–9 % of the total PNC, corresponding to 610–860 # cm⁻³. Polar plots indicate high contributions under NE winds at all sites except A, where the source appears to have a predominantly local origin (Fig. 8 and S9). The NE origin could be attributed to the same industrial and traffic sources described above.

3.5.4. Spatial and directional analysis of aircraft-related sources

To further validate the identification and spatial behaviour of aircraft-related sources, we examined how PNSDs and source contributions varied across the four monitoring sites depending on wind direction. Specifically, we compared the full dataset (all wind directions) with subsets filtered to include only hours when the wind came from the direction of the airport (between 55° and 75° for sites A and B, and

235°–275° for sites C and D).

PNSDs showed differences when filtering for wind from the airport direction (Fig. S10). For sites B, C, and D, although the peak of the PNSD during airport-wind periods shifted slightly toward coarser diameters, the PNSD was characterised by higher PNC from the PNSD peak to the finer sizes, suggesting a higher dominance of particles in the nucleation mode, which is consistent with freshly emitted aircraft-related particles. In contrast, under all wind directions, PNSDs tended to have higher concentrations in the Aitken and accumulation modes, indicating a broader mixture of sources. At the rural site D, the airport-wind PNSD showed a smoother decline, while the all-directions PNSD displayed a small secondary peak around 80 nm, with a less pronounced decrease in PNC beyond this point, suggesting influence from regional background sources.

Sources apportionment results reflect similar patterns (Fig. S11). At sites B, C, and D, the combined contribution of the two aircraft-related factors (*Takeoff* and *Taxiing*) increased during periods when the wind originated from the airport direction compared to the full dataset. At sites A and B (located southwest of the airport), the aircraft-related contributions increased from 28.6 % to 50.1 %–28.2 % and 67.7 %, respectively. At sites C and D (northeast of the airport), the increase was from 53.5 % to 34.5 %–83.6 % and 58.1 %, respectively. These directional increases support the attribution of these factors to airport activity, as they align with both prevailing wind directions and the distinct PNSD changes observed under those conditions. Furthermore, the increases were smaller at sites A and B than at C and D, likely because dominant winds blow from the SW, meaning that the northeastern sites are more frequently downwind of the airport and thus more affected by its emissions.

3.6. Limitations

Several limitations should be considered when interpreting the findings presented here, particularly regarding the temporal and spatial coverage of the data and the inherent assumptions behind each of the applied analytical methods. The two-month duration of the campaign, although supported by high time resolution measurements (5-min intervals), restricts the representativeness of the results concerning the averaged source contributions. Seasonal variation in meteorology, background pollution, and airport activity levels could significantly influence PNC and source contributions, especially at the more distant urban and rural monitoring sites. While the current dataset provides valuable insights into the impact of airport operations, a year-long campaign would be necessary to obtain more representativity of the quantitative results.

Uncertainty also varies depending on the method and proximity to the source. Source apportionment based on PNSD, which distinguishes taxiing from takeoff emissions, is considered to have relatively low uncertainty. These profiles were derived from measurements made near the emission sources, where meteorological influences are minimal and the signal is clearer. For this reason, the characterization of PNSD profiles for airport sources is regarded as one of the most robust results of the study.

In contrast, the estimation of the airport contribution to UFP concentrations based on the difference between upwind and downwind sites involves a number of assumptions that introduce potential sources of error. This additive model assumes that background sources contribute equally to both upwind and downwind sites and that particles emitted by the airport remain stable during transport. However, aerosol processes such as dilution, deposition, or chemical transformation can lead to a decrease in particle concentrations over distance. Furthermore, aircraft operating at low altitude may influence measurements even at sites classified as upwind, causing an underestimation of the airport's net impact. These factors introduce uncertainty into the absolute contribution values, particularly for the more distant sites.

The linear regression model adds further context by evaluating the

temporal variation of UFP concentrations as a function of explanatory variables such as the number of takeoffs and landings, wind parameters, and boundary layer height. The model showed good performance at sites directly downwind of the main runway, explaining 60 % and 51 % of the variance at sites C and D, respectively. These results can be interpreted with high confidence. In contrast, the model explained only 9 % and 32 % of the variance at sites A and B, likely due to a more complex mixture of sources, greater influence from local road traffic, and reduced exposure to aircraft emissions. Confidence in the model results at these upwind or more traffic-impacted sites is therefore lower.

4. Conclusions

This study assessed the impact of aircraft emissions on atmospheric ultrafine particle (UFP) concentrations near Brussels Airport, using high-resolution particle number size distribution (PNSD) data collected over a two-month period in autumn 2015 from four sites aligned with the airport's main runway. This monitoring design, covering both downwind and upwind positions, enabled a novel and robust assessment of airport-related influences on ambient UFP levels under different meteorological conditions.

Aircraft operations were shown to significantly impact air quality, with notable contributions to UFP concentrations observed up to 7 km downwind of the airport, especially for the 10–20 nm particle size class (smallest particles measured in this study), which constituted 65 % of total particle number concentrations (PNC) near the airport. Closer to the airport (within 750 m), average contributions from aircraft emissions to this size class were estimated at 15000–20000 #/cm³, peaking (30-min) at 65000–80000 # cm⁻³ during high-activity events. Even at distances of 5–7 km, 30-min peak contributions ranged from 4000 to 9000 # cm⁻³. While the overall UFP concentrations (both bulk PNC and airport contributions) declined with distance, the airport's influence remained discernible even at the furthest monitoring location. However, the two-month monitoring period limits the representativeness of long-term exposure, and longer measurement periods are needed.

Analysis of airport activity confirmed that PNCs correlated closely with landing and takeoff (LTO) operations, with significant impacts observed under favourable meteorological conditions. An attempt was made to distinguish the PNSD profiles of landing and takeoff, but the resulting distributions were very similar, likely due to the co-occurrence of both operations within the 5-min PNSD measurement intervals or an actual similarity in their emission characteristics.

Source apportionment using receptor modelling identified six factors contributing to UFP-PNSD, with taxiing and takeoff accounting for up to 64 % of PNC near the airport and 30 % at more distant sites. These two factors were well-resolved based on their modal diameters: takeoff emissions peaked around 20 nm, while taxiing emissions showed a smaller mode near 12 nm, consistent with differences in engine power settings. In total, airport sources contributed 6500–14000 # cm⁻³ at sites closest to the airport and 2300–2600 # cm⁻³ at distant sites. Given the proximity to the emission source and the distinct patterns in PNSD that are unique to each factor, these apportionment results are considered highly robust. Road traffic-related factors dominated further from the airport, while biomass burning and regional background sources made smaller contributions, though more pronounced at night.

Overall, this study highlights the substantial role of aircraft emissions in influencing UFP concentrations near Brussels Airport, with discernible effects on air quality extending several kilometres away. It provides a detailed-size resolved assessment of aircraft-related contributions. The use of a four-site setup along the runway allowed for a unique spatial characterization of airport impacts and the identification of distinct aircraft-related processes. The findings underscore significant regional air quality and public health implications, and support the need for targeted mitigation strategies.

CRedit authorship contribution statement

Meritxell Garcia-Marlès: Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Jan Peters:** Writing – review & editing, Writing – original draft, Methodology, Data curation, Conceptualization. **Jo Van Laer:** Writing – review & editing. **Evelien Frijns:** Writing – review & editing. **Patrick Berghmans:** Writing – review & editing. **Jeroen Staelens:** Writing – review & editing. **Jelle Hofman:** Writing – review & editing. **Thierry de Vos:** Writing – review & editing. **Olivier Brasseur:** Writing – review & editing. **Martine Van Poppel:** Writing – review & editing, Methodology, Data curation, Conceptualization. **Andrés Alastuey:** Writing – review & editing, Writing – original draft, Data curation, Conceptualization. **Xavier Querol:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2025.121375>.

Data availability

Data will be made available on request.

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4.2.5 Paper V

Effect of dispersion normalisation on long-term source apportionment and trends of ultrafine particles in Barcelona (NE Spain)

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Effect of dispersion normalisation on long-term source apportionment and trends of ultrafine particles in Barcelona (NE Spain)

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Highlights

- Dispersion normalisation improves ultrafine particle (UFPs) source apportionment.
- Road traffic dominates particle number concentrations (PNCs) in Barcelona.
- Dispersion-corrected analyses show higher photonucleation PNCs than raw data.
- Long-term trends show decreases in traffic-related UFPs, matching emission controls.
- COVID-19 sharply reduced traffic and photonucleation loads, probably from shipping.

Keywords

Particle number size distributions, Positive matrix factorization, Air quality, Ventilation coefficient, Inter-annual variability

Abstract

Ultrafine particles (UFPs, ≤ 100 nm) are a major component of urban air pollution, influenced by source emission rates (of UFPs and precursors) and meteorological conditions, both of which vary over time. This study evaluates the effect of dispersion normalisation on UFP source apportionment and long-term trends in the urban background of Barcelona (NE Spain) over a 12-year period (2013–2024). Hourly particle number size distributions (PNSD) were normalised using the ventilation coefficient (VC), derived from boundary layer height and wind speed, and analysed with positive matrix factorisation (PMF) for cold (October–March) and warm (April–September) seasons. Seven factors were identified, with road traffic as the dominant contributor. Dispersion corrected (DC) traffic contributions averaged 57% of particle number concentrations (PNCs), compared with 64% in the uncorrected (UC) analysis, indicating overestimation of local sources when dispersion is not considered. Photonucleation and Nucleation growth showed higher DC contributions (37% compared with 31% for UC), consistent with midday occurrence under favourable mixing conditions. Trend analysis revealed significant decreases in PNCs (-4.4% yr⁻¹), strongest in the Aitken mode, likely

linked to reduced traffic emissions following EURO 5/6 regulations, the introduction of diesel particulate filters (DPFs), and the implementation of Barcelona's low emission zone (LEZ). Traffic-related factors declined by -4.6% yr^{-1} , with the largest decrease in the diesel-related source. Nucleation growth and Regional-1 also declined, while Photonucleation and Regional-2 showed no clear trend. These findings highlight the value of dispersion normalisation in source apportionment and provide strong evidence of the effectiveness of emission control-policies in reducing traffic-related UFPs.

1. Introduction

Particulate matter (PM) significantly influences urban air quality worldwide and has substantial effects on human health. Evidence indicates that it contributes to respiratory and cardiovascular diseases, among others, and is associated to increased mortality (WHO, 2024).

PM is often quantified by mass concentration (in terms of mass per unit volume, $\mu\text{g m}^{-3}$). On the other hand, ultrafine particles (UFP), defined as particles with diameters less than 100 nm, contribute minimally to the PM mass concentrations, but comprise a large proportion of the total particle number concentration (PNC, measured in particles per cubic centimetre, $\# \text{ cm}^{-3}$). Given that UFPs generally contribute around 80% of the total PNC (Baldauf et al., 2016; Sun et al., 2019; Trechera et al., 2023), the term "PNC" without an upper size limit is commonly used as a surrogate for UFP.

Particle number size distribution (PNSD) is determined by measuring PNC for specific size ranges and are commonly categorised into three size modes: nucleation (<25 nm), Aitken (25–100 nm) and accumulation (100–1000 nm) (Seinfeld and Pandis, 2016). Although UFP have not specific air quality standards in the European Union (EU), neither specific WHO (2021) air quality guidelines, the new EU ambient air quality directive (EC, 2024) requires both UFP and PNSD measurements to be implemented in air quality monitoring supersites.

As recognised by WHO (2021), the body of evidence on the health effects for UFPs has grown but results are still inconsistent. Several studies suggest that the small size of UFPs allows them to reach the deeper parts of the respiratory system, translocating through the alveoli and reaching the circulatory system and, from there, any organ in the body (Oberdörster et al., 2005; Peters et al., 2006; Kreyling et al., 2014; Cassee et al., 2019). Oberdörster et al. (2004) also evidenced that UFPs might directly reach the brain by translocating through the olfactory bulb. Furthermore, exposure to UFPs has been linked to an increased risk of premature mortality (Ibald-Mulli et al., 2004; Tobías et al., 2018; Schwarz et al., 2023).

PNSDs can vary significantly across space and time, and can be categorised into distinct size modes associated with specific sources. Source apportionment studies in urban areas point to road traffic as a major source of UFPs (Pey et al., 2009; Dall'Osto et al., 2012; Harrison et al., 2011; Kumar et al., 2014; Brines et al., 2015; Beddows and Harrison, 2019; Rivas et al., 2019; Hopke et al., 2022; Kalkavouras et al., 2024, Garcia-Marlès et al., 2024b), accounting for more than 70% of the annual UFP ambient concentrations in most urban sites (Hopke et al., 2022; Garcia-Marlès et al., 2024b). UFPs from vehicles can be formed in the engine or in the atmosphere immediately after exhaust emission as these dilute and cool (Harrison et al., 2011; Rönkkö and Timonen, 2019). Typically, two major traffic-related particles modes are identified: around 30–35 nm, associated with spark-ignition (gasoline) engines, and 60–80 nm, linked to diesel engine emissions (Ogulei et al., 2007; Liu et al., 2014; Hopke et al., 2022; Vörösmarty et al., 2024). Additionally, finer particles are also associated with the nucleation of particles formed during the dilution of semi-volatile organic compounds (SVOCs) present in diesel exhaust (Harrison et al., 2011; Damayanti et al., 2023). In addition to the traffic-related emissions, photochemical nucleation from other gaseous precursors also contributes significantly, leading to new particle formation (NPF) events. These events are characterised by PNSDs peaking at the finest sized and are enhanced by high insolation and wind speed (WS), low relative humidity, low condensation sink (CS), and available precursor gases such as SO₂, NH₃, and amines, among the most relevant (Kulmala and Kerminen, 2008). Other contributions to PNCs include aircraft emissions, shipping, and domestic and regional sources (Stacey et al., 2020; Merico et al., 2020; Kuye et al., 2023).

Beyond the sources and formation mechanisms, PNCs are also strongly influenced by meteorological processes, with dispersion (both horizontal and vertical) typically leading to the dilution of particles, while stagnation to particle accumulation. Thus, when evaluating source contributions, strong dispersion can lead to underestimated values of specific emissions (Chen et al., 2022). Therefore, it is important to assess the long-term sources of PNCs after accounting for and minimising the impact of meteorological dilution of UFPs. This dilution is largely driven by solar radiation, which influences the planetary boundary layer height (BLH), typically higher during warm seasons and shallower during cold seasons, as well as by WS and precipitation (scavenging in this case) (Andronache, 2004; Kumar et al., 2011; Wang et al., 2023). Thus, dispersion normalisation has been recently shown to be an effective approach to reduce this variability. It is based on the ventilation coefficient (VC), determined by the BLH and WS (Ashrafi et al., 2009), which has led to improved source apportionment results (Dai et al., 2020; Chen et al., 2022; Hopke et al., 2024; Vörösmarty et al., 2024).

The aim of this study is to conduct a comprehensive source apportionment analysis of UFP in Barcelona (NE Spain), using a unique long-term dataset covering 12 years from 2013–2024, and to apply trend analysis to the source apportioned PNCs. A key objective is to account for the influence of meteorology-driven dispersion on UFP by applying a normalisation method based on the VC, and to compare the results obtained with and without this normalisation. Seasonal variability is also explored by analysing differences between cold and warm periods, to better understand UFP behaviour under varying atmospheric conditions (Hopke et al., 2024). This work provides new insights into the role of meteorological dispersion in shaping UFP source contributions and presents the first long-term dispersion normalised source apportionment and trend analysis of UFPs in an urban Mediterranean environment.

2. Methodology

2.1. Monitoring site and instrumentation

This study is based on data from an air quality supersite in Barcelona (northeastern Spain), located on the western Mediterranean coast. Measurements were conducted at the urban background site of Palau Reial (41.39° N, 2.12° E; 80 m a.s.l.), located in a residential area in northwestern Barcelona. Since 2021, Barcelona Palau Reial site has been part of the Aerosol, Clouds and Trace gases Research Infrastructure (ACTRIS) national facilities (NF). The site is influenced by road traffic emissions from one of Barcelona's busiest avenues, located approximately 200 m away, with an average daily traffic volume of 60000 vehicles/working day (Ajuntament de Barcelona, 2021). In addition to road traffic, the site is also influenced by numerous industrial facilities and three power plants surrounding the city, by the Port of Barcelona (6 km south of the monitoring site), one of the busiest harbours in the Mediterranean Sea, by Barcelona-El Prat Airport (10 km southwest of the site), which recorded 350000 operations in 2024 (AENA, 2025), by large agricultural and farming areas in the region, and by the domestic and commercial sources.

The data cover a continuous 12-years observation period, from 2013 to 2024. PNSD measurements, covering 103 size bins with midpoint diameters from 12.2 to 478.3 nm, were obtained using a scanning mobility particle sizer spectrometer (SMPS) TSI 3080 and a condensation particle counter (CPC) TSI 3772 for the period 2013–2019, and a SMPS TSI 3082 and CPC TSI 3750 for the period 2020–2024. Particulate matter (PM₁₀ and PM_{2.5}) and gaseous pollutants (SO₂, NO, NO₂, O₃, and CO) concentrations were also monitored at the site. Meteorological parameters (temperature (T), pressure (P), relative humidity (RH), radiation (RAD), rain (R), wind direction (WD), and WS) were obtained from the Faculty of Physics of the University of Barcelona, located approximately 300 m from the

monitoring site. All instrumentation complied with European standards. The data were then averaged to hourly values.

Hourly BLH data were retrieved from the ERA5 atmospheric model (Hersbach et al., 2023), the fifth generation European Centre for Medium-range Weather Forecasts (ECMWF) reanalysis for the global climate and weather from 1940 onwards, produced by the Copernicus Climate Change Service (C3S).

2.2. Dispersion normalisation

Dispersion normalisation has been applied to the data to reduce the effects of meteorological variation on PNCs using the VC, an index that represents the potential volume of air available per unit time for the dilution of pollution released into the atmosphere. For each measurement time period i , VC_i is defined as:

$$VC_i = BLH_i \times \bar{u}_i$$

where BLH_i is the boundary layer height, and $\bar{u}_i$ is the mean WS, for the time period i (Ashrafi et al., 2009). Concentration values (PNC and mass concentrations) C_i are then multiplied by the ventilation coefficient ratio (VC_{ratio}) of the VC_i over the average VC for the whole sampling period:

$$C_{Vi} = C_i \times \frac{VC_i}{\overline{VC}}$$

where C_i is the measured concentration for the time period i , C_{Vi} is the corresponding normalised concentration, and $\overline{VC}$ is the average VC. In this study, $\overline{VC}$ was $1021 \text{ m}^2 \text{ s}^{-1}$.

After performing the source apportionment with the dispersion normalised data, the resulting source contributions were unnormalised by dividing the resolved values by the VC_{ratio} to obtain the real concentrations.

2.3. Positive matrix factorisation

Source apportionment was performed using positive matrix factorisation (PMF; Paatero and Tapper, 1994), the most recently used data analysis method to identify and apportion the sources of PNSD. PMF is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into a factor profile matrix and a factor contribution matrix. These matrices require interpretation to identify source types using measured source profile information and emissions inventories. Given that the current implementation of the U.S. Environmental Protection Agency (US EPA; Norris et al., 2014) PMF 5.0 program does not handle datasets of more than 500000 data points, the tool developed by

Hopke et al. (2023), which uses the multilinear engine 2 (ME-2; Paatero, 1999), was employed in this study.

Hourly PNSD data (103 geometrically size bins) were combined with the hourly concentrations of ancillary pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO) to help identify the sources. Uncertainties in the PNC values were estimated using the approach of Ogulei et al. (2007) and Rivas et al. (2019):

$$s_{ij} = \alpha_{ij} + C_3 \cdot n_{ij}$$

where $\alpha_{ij} = 0.01(n_{ij} + \bar{n}_{ij})$, n_{ij} is the observed concentration of sample i in size bin column j , $\bar{n}_{ij}$ is the arithmetic mean of the observed concentration in size bin j , and C_3 is a constant determined by trial-and-error. Uncertainties for the 3 % lowest and 3 % highest size bins were multiplied by a factor of 2, and by a factor of 1.5 for the subsequent 3 % lower and 3 % higher size bins to reduce the increased measurement error of the lowest and highest bins of the PNSD (Wiedensohler et al., 2017; Rivas et al., 2019). Uncertainties for the ancillary pollutants were determined by Polissar et al. (1998) equation. An additional uncertainty K was applied to all the variables to optimise the model. Values of C_3 and K for each PMF are presented in Table S1.

The resulting dataset comprised 63813 hourly values of 111 variables, in total 7.083.243 data points. As noted in Hopke et al. (2023), the ME-2 program also has a computational limit on the number of equations it can process, which represents a limit of less than 6 million data points. Consequently, it was not feasible to analyse the entire data set in a single analysis. Given that Zhou et al. (2004) and Ogulei et al. (2007) reported seasonal variations in the size distribution profiles by changes in atmospheric chemistry, the dataset was divided into two subsets: a cold season (October, November, December, January, February, and March) and a warm season (April, May, June, July, August and September). PMF modelling was then conducted separately for each period, aggregated across all 12 years. For each seasonal subset, PMF was run twice: once with dispersion normalisation applied (dispersion corrected source apportionment, hereafter DC-SA) and once without (uncorrected source apportionment, UC-SA) to allow for comparison. This resulted in a total of four PMF analyses.

For each dataset, PMF was executed multiple times with varying number of factors to determine the optimal solution, and the corresponding factor displacement (DISP) was also assessed. The best solutions were chosen based on accepted criteria (Belis et al., 2019; Hopke et al., 2023): the scaled residuals are randomly distributed preferably between -3 and +3, the $Q_{\text{true}}/Q_{\text{exp}}$ ratio is close to 1 (see Table S1), and results have the most physically meaningful interpretation. Source profiles and contributions were evaluated with the R statistical software (v4.4.2, R Core Team, 2024) and the package *Openair* (Carslaw and Ropkins, 2012).

2.4. Trend analysis

Data series in Barcelona were evaluated to detect monotonic trends over the 12-year study period using the Theil-Sen estimator (Sen, 1968; Theil, 1992), implemented via the *Openair* package in R (Carslaw and Ropkins, 2012). This non-parametric method provides a robust estimate of linear trends, with reduced sensitivity to outliers. The analysis was applied to hourly PNCs, the three size modes (nucleation, Aitken, and accumulation), various pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO), and source contributions to PNCs. Monthly averages were calculated using a minimum data coverage threshold of 30% of the hourly measurements. The magnitude of the trends was quantified using the Theil-Sen slope (i.e., the annual change in concentration), along with associated confidence intervals and statistical significance (ss), providing insight into whether concentrations have increased, decreased, or remained stable over the study period.

The data analysed includes source contributions derived from PMF, which include inherent variability and negative values due to source apportionment uncertainties. Because of this, attempts to apply seasonal decomposition to de-seasonalise the data were unsuccessful due to convergence issues, attributable to irregular seasonal patterns and the complex, mixed nature of the source contributions. Consequently, de-seasonalisation was not applied to ensure stability and robustness of results. Sensitivity analyses indicated that the exclusion of de-seasonalisation did not substantially alter the main trend conclusions.

3. Results and discussion

3.1. Mean concentrations and influence of dispersion correction

Annual mean PNCs in Barcelona showed a general decrease over the 12-year study period (Table S2). In 2013, the mean PNC was 11900 # cm⁻³, followed by a moderate decrease in the subsequent years, reaching 10400 # cm⁻³ in 2019. A significant reduction occurred in 2020 most probably due to COVID-19 lockdown measures, with concentrations dropping to a minimum of 6200 # cm⁻³ (as an annual average). In the post-pandemic years, PNC levels partially rebounded but below pre-pandemic values, reaching 8400 # cm⁻³ in 2024. The trend analysis is presented in subsequent sections.

Annual variations in PNCs are influenced not only by changes in emissions but also by meteorological conditions affecting pollutant dispersion. To account for this, normalised annual mean PNCs were also calculated using the VC_{ratio}, as described in the methodology section, to allow for a more robust comparison of mean concentrations across years. The DC means followed similar temporal patterns but were generally lower, with values of 10700, 8600, 5500, and 7300 # cm⁻³ in 2013, 2019, 2020, and 2024, respectively (Table S2).

To better understand the meteorological influences driving the variability of UFPs, the diurnal and monthly patterns of WS, BLH and VC_{ratio} were analysed. Mean diurnal variations of all three parameters exhibited very similar patterns in both warm and cold seasons, with a broad peak at midday reflecting enhanced atmospheric mixing, but also with high WS at night during the cold season (Figure S1). The VC_{ratio} exceeded 1 (up to 2.1) between 8:00 and 17:00 UTC, indicating stronger dispersion conditions during the day, while it dropped below 1 (down to 0.3) during the rest of the day, reflecting more stagnant conditions typical of the night-time. This pattern highlights the pronounced diurnal cycle of dispersion and supports the use of the VC_{ratio} to normalise PNCs and reduce meteorological effects.

Monthly variations show higher WS during the cold season due to increased temperature contrasts and stronger pressure gradients, while calmer conditions prevail in the summer months (Figure S2). BLH exhibited higher values in spring and autumn, and lower values in both summer and winter. Although increased solar radiation in summer typically enhances surface heating and consequently stronger convective processes (Moreira et al., 2020), the lower BLH values observed during this season are attributed to the cooling effect of sea-breeze incursions, which limit vertical mixing (Sicard et al., 2005). As a result, atmospheric dispersion is generally poorer in the warm season, as reflected in the lower values of the VC_{ratio} .

The impact of dispersion normalisation on PNCs is showed in Figure 1, which compares UC and DC hourly mean PNCs separately for the cold and warm seasons. In both seasons, a similar pattern is observed. After correction, the typical traffic-related peaks during morning and evening rush hours are very mild, while a very pronounced peak is observed around midday.

This suggests that the original diurnal variation was largely driven by meteorological dilution rather than actual changes in emissions. However, the corrected pattern should be interpreted with caution, as it may be influenced by uncertainties in the modelled BLH data and the estimation of the VC_{ratio} , both of which are subject to limitations and assumptions inherent in their calculation.

3.2. Source apportionment

3.2.1. Identification of the factors

Based on the criteria described in the methodology section, a six-factor PMF solution was selected for all four analyses: cold season (DC-SA and UC-SA) and warm season (DC-SA and UC-SA). Profiles were firstly interpreted for both seasons as *Traffic-1*, *Mixed traffic*, *Traffic-2*, *Photonucleation*, *Regional-1*, and *Regional-2*, coincident with Garcia-Marlès et al. (2024b). However, during the warm season, a factor (*Traffic-1*) with a size mode between 25–30 nm exhibited

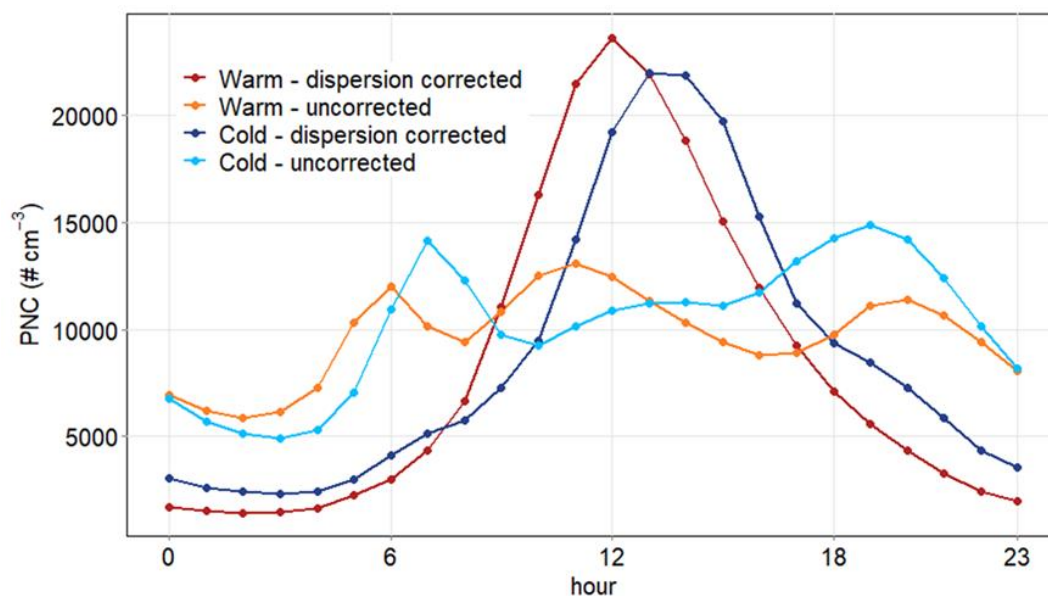


Figure 1. Seasonal mean diurnal variations of dispersion corrected and uncorrected particle number concentrations (PNC), during warm season (April–September) and cold season (October–March). Values represent hourly means averaged over the full period.

peaks during the morning and evening rush hours (associated with road traffic emissions) and around midday (attributed to photonucleation processes). Since PMF was unable to separate these contributions due to their similar PNSDs, the factor was subsequently split into two sources using a methodology developed by Rodríguez and Cuevas (2007) and Rivas et al. (2019). To apply this method, NO_2 concentrations were used as a proxy for traffic emissions. The 10th percentile of the ratio between the factor and NO_2 (scaling factor) was calculated using data corresponding to the morning traffic peak (assuming most particles would be from traffic). The selection of the 10th percentile was based on previous studies (Reche et al., 2011; Kulmala et al., 2016; Kalogridis et al., 2018; Nie et al., 2022). This scaling factor was applied to hourly NO_2 concentrations to estimate the traffic contribution (*Traffic-1*), while the photonucleation component was obtained by subtraction (named *Nucleation growth*). Thus, the warm season included seven factors instead of six.

Results from the DC-SA and UC-SA showed similar source profiles and temporal patterns for both the cold and warm seasons, with differences mainly in their relative contributions (discussed in the next section). Figure 2 presents the PNSD profiles and diurnal and weekly patterns for both seasons for the DC-SA, while Figure 3 shows the corresponding polar plots. Equivalent plots for the UC-SA, which displayed very similar characteristics, are provided in Figures S5–S8. Contributions to the

variance of PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO for the four analyses can be found in Tables S3–S6.

Road traffic sources

Three road traffic-related factors were identified in both the cold and warm seasons, showing distinct PNSD profiles within the Aitken mode but similar temporal patterns. The three factors exhibited peaks during rush hours in the morning and evening and showed lower PNCs on weekends (Figure 2), consistent with typical traffic temporal patterns in Barcelona (Ajuntament de Barcelona, 2025). These contributions were lower during the warm season, with August showing the lowest PNCs (Figure S9), when the car fleet decreases on holidays.

Traffic-1 was characterised by size mode peaks at 27–29 nm (Figure 2) and was associated with low but notable contributions of NO₂ and CO (Tables S3–S6). As mentioned above, this factor was split into two sources for the warm season. For the cold season, this approach was not applied because the midday peak was too low. However, it should be noted that, in the cold period, this factor may also include particles generated from photochemical processes. This factor mainly originated locally, predominating at low WS (Figure 3). High contributions observed from the south during the cold season are attributed to the photonucleated particles that were not separated from this factor. Given these features, this factor is attributed to gasoline vehicle emissions (Ogulei et al., 2007; Liu et al., 2014; Hopke et al., 2022) and nucleation of particles generated during the dilution of diesel exhaust SVOCs (Harrison et al., 2011, Damayanti et al., 2023). Some studies have attributed similar profiles to freshly emitted traffic particles on nearby roads (Gu et al., 2011; Rivas et al., 2019).

Traffic-2 was characterised by size mode peaks at 88–98 nm (Figure 2) and exhibited higher contributions of NO₂ and CO, with additional contributions to NO, SO₂, PM₁₀, and PM_{2.5} (Tables S3–S6). Polar plots indicated a local origin, with elevated concentrations at low WS (Figure 3). This factor is associated with primary diesel vehicle emissions (Ogulei et al., 2007; Liu et al., 2014; Hopke et al., 2022), mostly from heavy- and light-duty vehicles (Zhang et al., 2020). Diesel emissions are responsible for most of the traffic-related NO_x emissions (He et al., 2015). Other studies suggest that this larger mode may result from particle coagulation as they move away from the emission source, i.e., aged traffic particles (Gu et al., 2011; Rivas et al., 2019).

The *Mixed traffic* factor fell within the size range between *Traffic-1* and *Traffic-2*, with mode peaks at 46–53 nm (Figure 2). It also exhibited significant contributions of NO₂, CO, and SO₂ (Tables S3–S6), and was primarily of local origin, although with notable PNC contributions from the south (Figure 3). This factor is attributed to particles emitted from vehicles with different fuel types, mixed with other urban sources (Garcia-Marlès et al., 2024b).

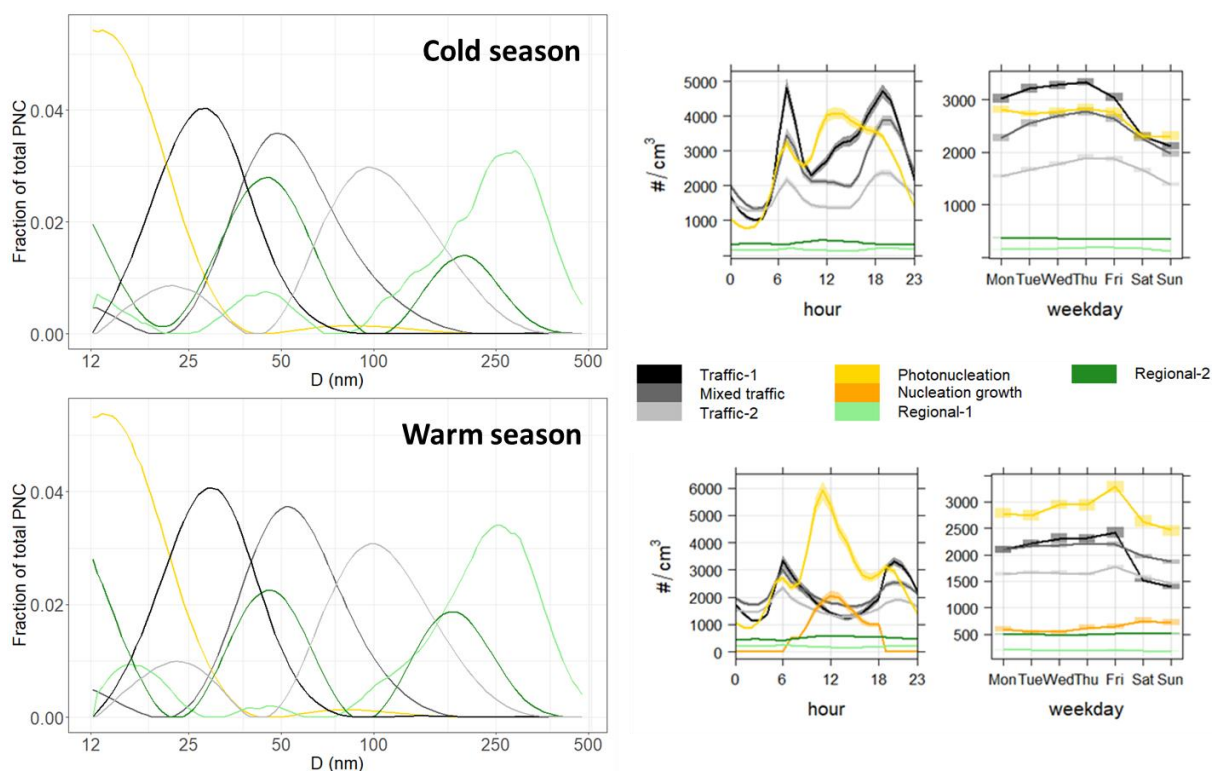


Figure 2. Particle number size distribution (PNSD) profiles (left), and diurnal and weekly patterns (right) of the sources identified in the dispersion corrected source apportionment (DC-SA) for the warm season (April–September, bottom) and the cold season (October–March, top). For the warm season, the profile initially identified as *Traffic-1* (black) was subsequently separated into two distinct sources: *Traffic-1* (black) and *Nucleation growth* (orange). Displacement values and the percentage of explained variation in the PNSDs are presented in Figures S3–S4.

Photonucleation sources

Photonucleation was characterised by size modes with peaks at 12–13 nm, with higher PNCs observed around midday, coinciding with peak solar radiation (Figure 2), and during the warm season (Figure S9). Lower peaks observed during the morning and evening rush hours may be attributed to NPF from the nucleation of particles immediately after the emission of SVOCs into the atmosphere from vehicle exhaust, as they dilute and cool. These processes arise mostly from the regeneration of diesel particle filters (DPFs) in diesel vehicles and from catalytic converters in gasoline ones (Harrison et al., 2011; Mamakos and Martini, 2011; Trechera et al., 2023; Saarikoski et al., 2024). This factor exhibited significant levels of SO₂ and O₃ (Tables S3–S6). It is associated with photochemically induced NPF events, which occur under conditions of high solar radiation and WS, and are enhanced by a low CS when traffic-related emissions decrease (McMurry and Friedlander, 1979; Kulmala and Kerminen, 2008; Brines et al., 2015; Salma et al., 2021). In Barcelona, it is mostly

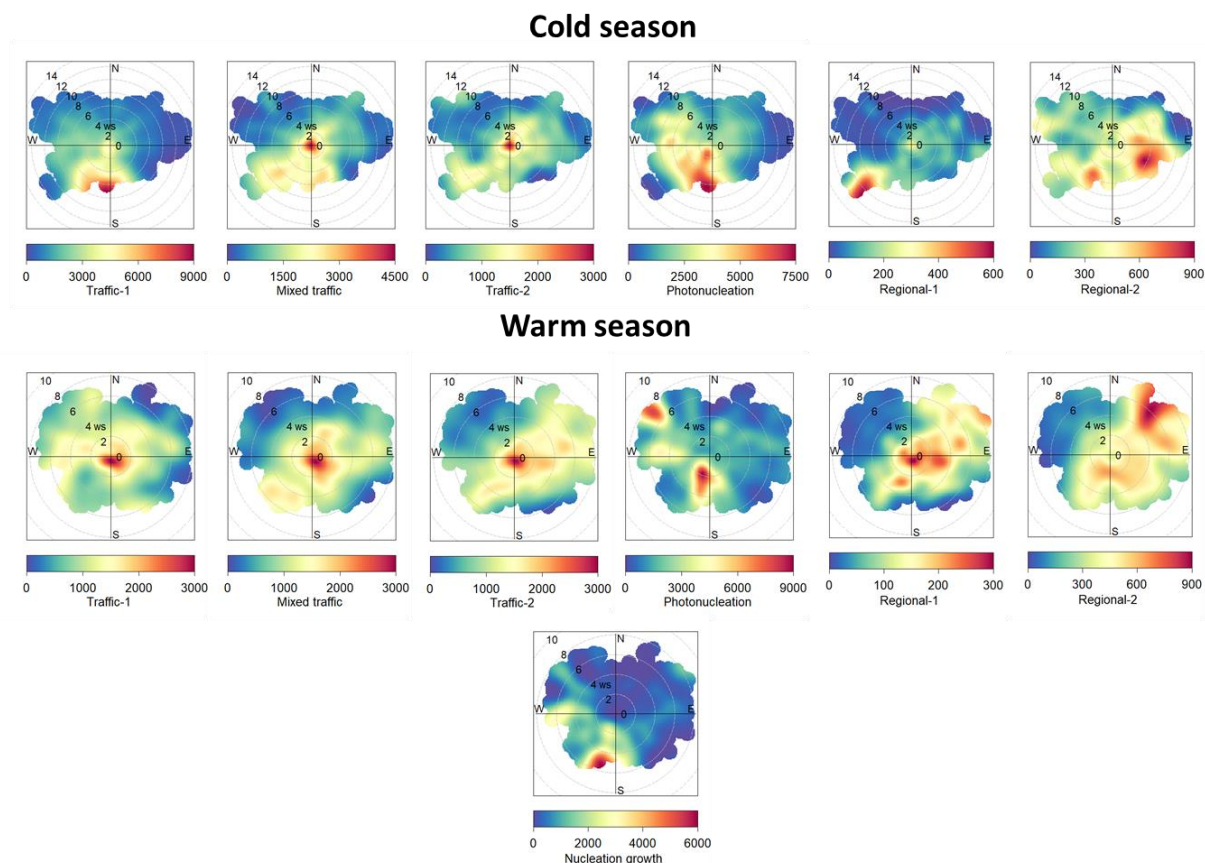


Figure 3. Polar plots showing the source contributions identified in the dispersion corrected source apportionment for the cold season (October–March, top) and warm season (April–September, bottom).

influenced by particles nucleating in high SO_2 plumes emitted from the harbour, as supported by the elevated contributions observed from the south (Figure 3), where the harbour is located. Contributions from the northwest may be attributed to industrial emissions of precursor gases such as SO_2 , NH_3 , and amines, which can also lead or favour to NPF events (Kulmala and Kerminen, 2008). The west origin is associated with aircraft emissions from Barcelona Airport, which have also been reported to contribute to nucleation mode particles (Mazaheri et al., 2013; Masiol et al., 2016; Tremper et al., 2022; Ridolfo et al., 2024; Garcia-Marlès et al., 2025).

The *Nucleation growth* factor was only identified during the warm season, which was extracted from the *Traffic-1* factor using the approach described above. This component is attributed to the growth of photonucleation particles, as indicated by its PNSD peak being shifted toward larger sizes (29 nm) compared to the *Photonucleation* source peak (13 nm). It exhibited a peak around midday, with the maximum occurring approximately one hour later than the *Photonucleation* factor (Figure 2), supporting the interpretation of particle growth (Brines et al., 2015). Polar plots revealed a similar

origin to *Photonucleation*, with high contributions mainly from the harbour (south), and from the airport (west) (Figure 3).

Regional sources

Two (mostly) regional sources were identified in both seasons, characterised by similar multimodal distributions with primary peaks in the accumulation mode and secondary peaks in the nucleation and Aitken mode, but exhibiting different temporal patterns.

Regional-1 was characterised by major size mode peaks at 255–290 nm, with lower peaks ranging from 12–45 nm (Figure 2). It exhibited higher PNCs at night, during traffic rush hours, and throughout the cold season (Figure S9). This factor was associated with notable contributions to PM₁₀ and PM_{2.5} (Tables S3–S6). It is attributed to secondary aerosols, mainly composed of nitrate particles formed via the atmospheric oxidation of NO_x, which is enhanced during the night and the traffic rush hours (Seinfeld and Pandis, 2016). The elevated PNCs observed at night are also consistent with semi-volatile nitrate condensing on locally emitted particles under conditions of lower temperature and higher relative humidity (Dall'Osto et al., 2009; Masiol et al., 2016). Polar plots indicated different origins of these particles (Figure 3). Locally, particles may arise from the gas-to-particle conversion of local NO_x emissions from vehicles. Contributions under high wind speeds may reflect from particles formed during the transport and reaction of precursor gases (Kasumba et al., 2009; Squizzato et al., 2019). The elevated contributions observed from the southwest may be associated with dust events originating from the Sahara Desert in Africa (Garcia-Marlès et al., 2024b).

Regional-2 was characterised by a bimodal distribution with peaks ranging between 33–46 nm and 162–200 nm (Figure 2). Contributions of this factor were higher around midday, and during the warm season (Figure S9). Very high levels of O₃, SO₂, PM₁₀, and PM_{2.5} were associated to this factor (Tables S3–S6). Polar plots showed similar distributions as O₃, with elevated contributions from the southwest and southeast during the cold season and from northeast during the warm season (Figures 3 and S10). This factor is associated with secondary aerosols, mainly sulphates and secondary organic aerosols, formed when photochemical activity is higher (Liu et al., 2014). The oxidation of SO₂ and VOCs during high summer insolation driven by O₃ produces NPF with a subsequent growth yielding to coarser particles (Ehn et al., 2014; Hopke et al., 2022). In the Western Mediterranean, the vertical recirculation of air masses is a major cause of regional O₃ pollution episodes, most favoured during warm months (Millán et al., 2000; Gangoiti et al., 2001; Carnerero et al., 2019).

3.2.2. Source contributions and impact of dispersion normalisation

Relative contributions of the identified sources to average PNCs for both the DC-SA and UC-SA are shown in Figure 4 for the warm and cold seasons. Although source profiles and temporal patterns were highly similar between the two approaches, the relative contributions differed substantially. Previous studies have also reported notable differences in source contributions when applying dispersion correction methods, highlighting the importance of accounting for meteorological variability when interpreting source apportionment results (Dai et al., 2020; Vörösmarty et al., 2023; Hopke et al., 2024).

In both seasons, total road traffic (the sum of *Traffic-1*, *Mixed traffic* and *Traffic-2*) was the major contributor to PNC in Barcelona, accounting for 60% and 53% of the average PNC during the cold and warm seasons, respectively, according to the DC-SA. High contribution from traffic emissions to PNC has also been reported in literature for many urban environments around the world (Pey et al., 2009; Dall'Osto et al., 2012; Liu et al., 2014; Beddows et al., 2015; Brines et al., 2015; Sowlat et al., 2016; Rivas et al., 2019; Squizzato et al., 2019; Vörösmarty et al., 2023; Kalkavouras et al., 2024, Garcia-Marlès et al., 2024b). Among the traffic-related factors, *Mixed traffic* and *Traffic-2* showed similar contributions across seasons, accounting for 19–22% and 13–14% of the average PNC, respectively. *Traffic-1* contributed notably more in the cold season (25%) than in the warm season (20%). This seasonal difference may be partly explained by the fact that, in the cold season, the *Traffic-1* factor could also include particles formed through photochemical processes, as the factor-splitting approach was not applied during this period, as previously discussed. This overlap may also help explain the higher overall contributions to road traffic during the cold season. Additionally, traffic volumes in winter are generally higher than in summer in Barcelona, further supporting the observed seasonal trend. In terms of PNC, the absolute concentrations of total road traffic for the DC-SA were 5300 and 4400 # cm⁻³ for the cold and warm seasons, respectively. For the UC-SA, total traffic contributions were even higher, accounting for 69% and 58% of the average PNC and during the cold and warm seasons, respectively, corresponding to absolute concentrations of 7000 and 5600 # cm⁻³. Despite these higher contributions, the seasonal trend for each traffic-related factor remained consistent with those observed in the DC-SA. Since traffic emissions are primarily local and strongly affected by dispersion conditions, the lack of correction can lead to an overestimation of their contribution consistent with the higher traffic contributions observed in the UC-SA, while the DC-SA accounts for these dispersion effects, providing source contributions that more accurately reflect emission strength rather than meteorological variability.

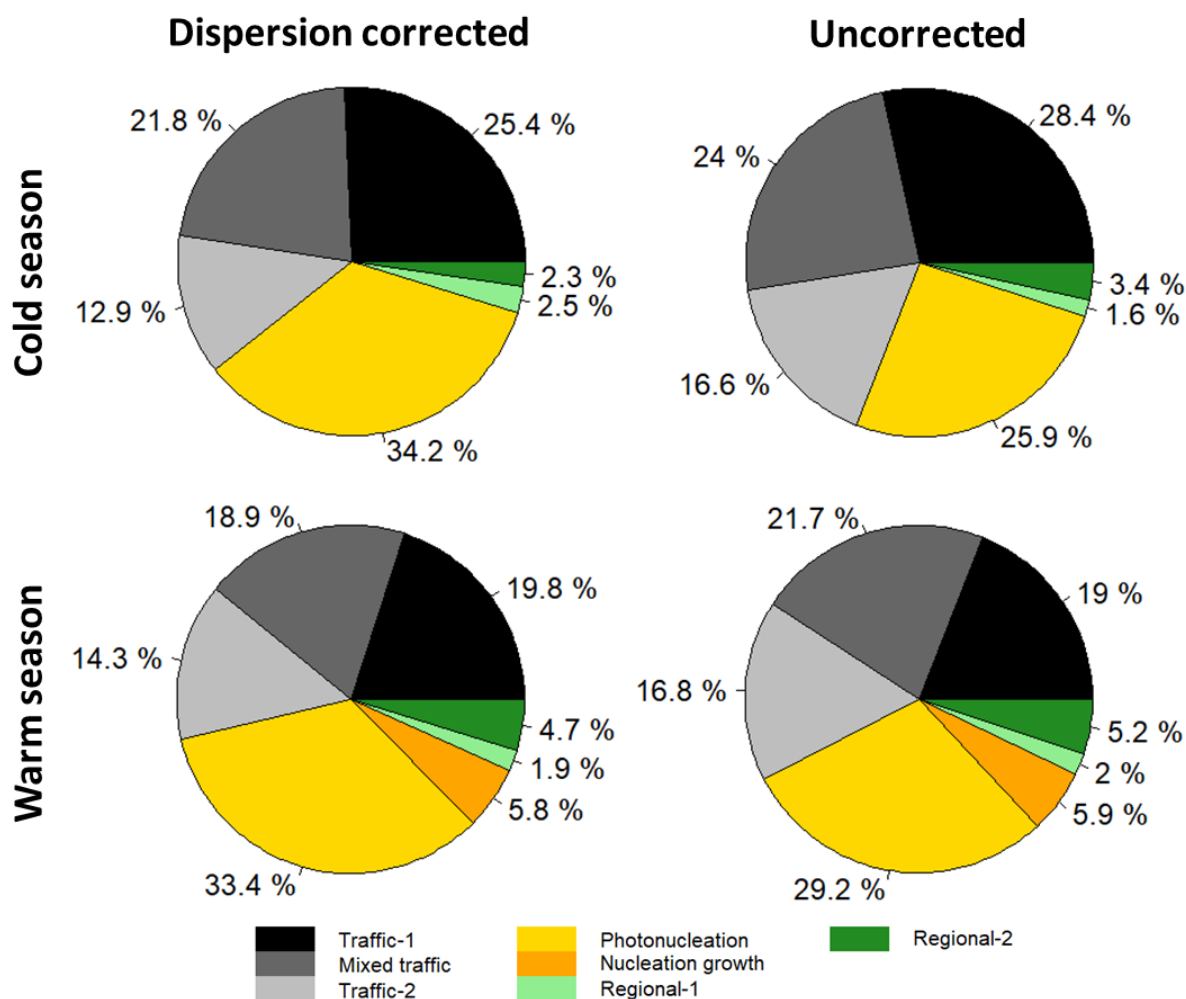


Figure 4. Relative contributions (%) of particle number concentration (PNC) from the sources identified in the dispersion corrected (left) and uncorrected (right) source apportionment for the warm season (April–September, bottom) and the cold season (October–March, top).

The dispersion correction proved particularly relevant for the *Photonucleation* source, which showed a marked increase in its contributions in the DC-SA. In the cold season, Photonucleation accounted for 34% of the average PNC in the DC-SA (corresponding to $3000 \text{ \# cm}^{-3}$), compared to 26% in the UC-SA ($2600 \text{ \# cm}^{-3}$). In the warm season, the combined contribution of *Photonucleation* and *Nucleation growth* reached 39% in the DC-SA ($3200 \text{ \# cm}^{-3}$) and 35% in the UC-SA ($3400 \text{ \# cm}^{-3}$). This enhancement in the relative contributions of the DC-SA is consistent with the fact that photochemical nucleation typically occurs around midday, when dispersion conditions are more favourable due to increased atmospheric mixing. Without correcting for dispersion, the UC-SA underestimates this source, as its contribution becomes diluted by the higher ventilation during the hours when nucleation is most active.

Lower contributions to the average PNC were observed for the regional factors, ranging from 2–3% for *Regional-1* and 2–5% for *Regional-2* across all source apportionment analyses. Due to their small contributions, the reliability of their interpretation is limited.

3.3. Inter-annual trends

Trend analysis results of PNCs, size modes, source contributions to PNC, and ancillary pollutants for the full-year, DC dataset are presented in Figures 5 and S11. Corresponding results for the UC dataset, as well as for the cold and warm seasons (both DC and UC), are provided in Figures S12–16. Detailed numerical results of the Theil-Sen slopes and confidence intervals are listed in Tables S7–12.

For both UC and DC datasets, the Theil-Sen monotonic slopes indicated consistent decreases across most of the variables analysed. After applying the DC, sharper decreases were observed for most of the variables analysed, but the same trends were obtained for the two analyses.

For the DC analysis, PNCs showed a ss 2013–2024 decrease of -439 $[-609, -285]$ $\# \text{ cm}^{-3} \text{ yr}^{-1}$, corresponding to a relative decrease of -4.4 $[-5.4, -3.1]$ $\% \text{ yr}^{-1}$. The three size modes also showed ss decreases, with slopes of -137 $[-224, -60]$, -249 $[-326, -179]$, and -50 $[-62, -40]$ $\# \text{ cm}^{-3} \text{ yr}^{-1}$ for the nucleation, Aiken, and accumulation modes, respectively, corresponding to -3.4 $[-4.7, -1.8]$, -5.0 $[-5.9, -4.0]$, and -4.8 $[-5.5, -4.2]$ $\% \text{ yr}^{-1}$. The strongest decrease was observed in the Aitken mode, most likely due to reduced emissions of road-traffic particles, which are typically found in this size range. These reductions are most probably associated with the implementation of European emission standards, particularly the requirement under EURO 5 and EURO 6 standards that diesel vehicles be equipped with DPFs (Damayanti et al., 2023). In contrast, the weaker decrease observed for nucleation mode particles may be linked to the limited abatement effect of DPFs on the finest UFP fraction, as these filters do not retain SVOCs emitted by diesel engines, which can nucleate and generate delayed primary particles (Chen et al., 2022; Damayanti et al., 2023). Alternatively, the trend may also reflect a decrease in the CS, which enhances the likelihood of NPF (Spracklen et al., 2010; Kulmala et al., 2017).

The meteorological analysis indicated a ss increase in T and RH, together with a decrease in WS, while no ss trends were found for the other variables (Figure S17). The warming trend is consistent with the weaker decrease observed in the nucleation mode, as higher T have been associated with reduced traffic-related UFP concentrations (Weichenthal et al., 2008), while at the same time they can modulate photochemical nucleation depending on concurrent RH, RAD, and CS conditions (Hamed et al., 2011; Yu et al., 2017; Brines et al., 2015; Tuovinen et al., 2021). Thus, the observed

decreases in nucleation mode particles may partly reflect the combined effect of emission controls and increasing ambient T, and low WS (the later favouring CS).

Additionally, the reductions in PNC observed in this study were sharper than those reported for 2013–2019 by Garcia-Marlès et al. (2024a), who found a ss slope of $-2.7 \text{ \% yr}^{-1}$ in the Aitken mode but no significant trends in the nucleation or accumulation modes. Extending the analysis period to 2013–2024 in the present study captured the pronounced decrease in PNCs associated with the COVID-19 lockdown measures in 2020–2021 (reducing markedly traffic, but also cruising), as well as the introduction of the low emission zone (LEZ) in Barcelona in 2020 and its further tightening in 2022, and the lower sulphur (S) content (from 3.5% to 0.5%) of shipping fuel requested by the International Maritime Organization (IMO) since January 2020 (IMO, 2020).

Seasonal analysis further supports the role of traffic emissions in driving the observed decreases. PNC reductions were more pronounced during the DC cold season ($-4.6 [-6.3, -2.7] \text{ \% yr}^{-1}$), when road traffic intensity is higher, compared with the DC warm season ($-4.0 [-5.3, -2.6] \text{ \% yr}^{-1}$), due to the lower one in July and August. This seasonal contrast was also evident in the Aitken and accumulations modes, which showed stronger decreases during the cold season.

Consistent with the decreases observed for the size modes, traffic-related source contributions to PNC also declined. Total traffic (the sum of *Traffic-1*, *Mixed traffic*, and *Traffic-2*) showed a ss decrease of $-395 [-475, -310] \text{ \# cm}^{-3} \text{ yr}^{-1}$, corresponding to a relative decrease of $-4.6 [-5.1, -3.9] \text{ \% yr}^{-1}$. This reduction was sharper than that reported by Garcia-Marlès et al. (2024b), who found a ss slope of $-3.4 \text{ \% yr}^{-1}$ for Total traffic during 2013–2019 in Barcelona. Among the three traffic-related factors identified, *Traffic-2* exhibited the largest decrease, with a ss slope of $-5.1 [-5.8, -4.4] \text{ \% yr}^{-1}$, followed by *Traffic-1* ($-4.7 [-5.5, -4.0] \text{ \% yr}^{-1}$) and *Mixed traffic* ($-4.2 [-4.9, -3.5] \text{ \% yr}^{-1}$). In terms of absolute PNC, this corresponds to decreases of $-125 [-153, -99]$, $-144 [-182, -122]$, and $-123 [-158, -93] \text{ \# cm}^{-3} \text{ yr}^{-1}$, for *Traffic-2*, *Traffic-1*, and *Mixed traffic*, respectively. The strongest decrease for *Traffic-2*, which is primarily associated with diesel vehicle emissions, is consistent with the implementation of EURO 5 regulations requiring diesel vehicles to be equipped with DPFs (Damayanti et al., 2023). In addition, the lowest contributions were observed during 2020–2021, reflecting the sharp reduction in road traffic activity associated with COVID-19 restrictions. In the subsequent years, traffic-related contributions increased again as mobility recovered, although they remained below pre-pandemic levels, most probably due to the implementation of the LEZ. These trends are also consistent with the concurrent decreases observed for traffic-related gaseous pollutants such as NO, NO₂, and CO, further supporting the role of emission controls in driving the observed reductions.

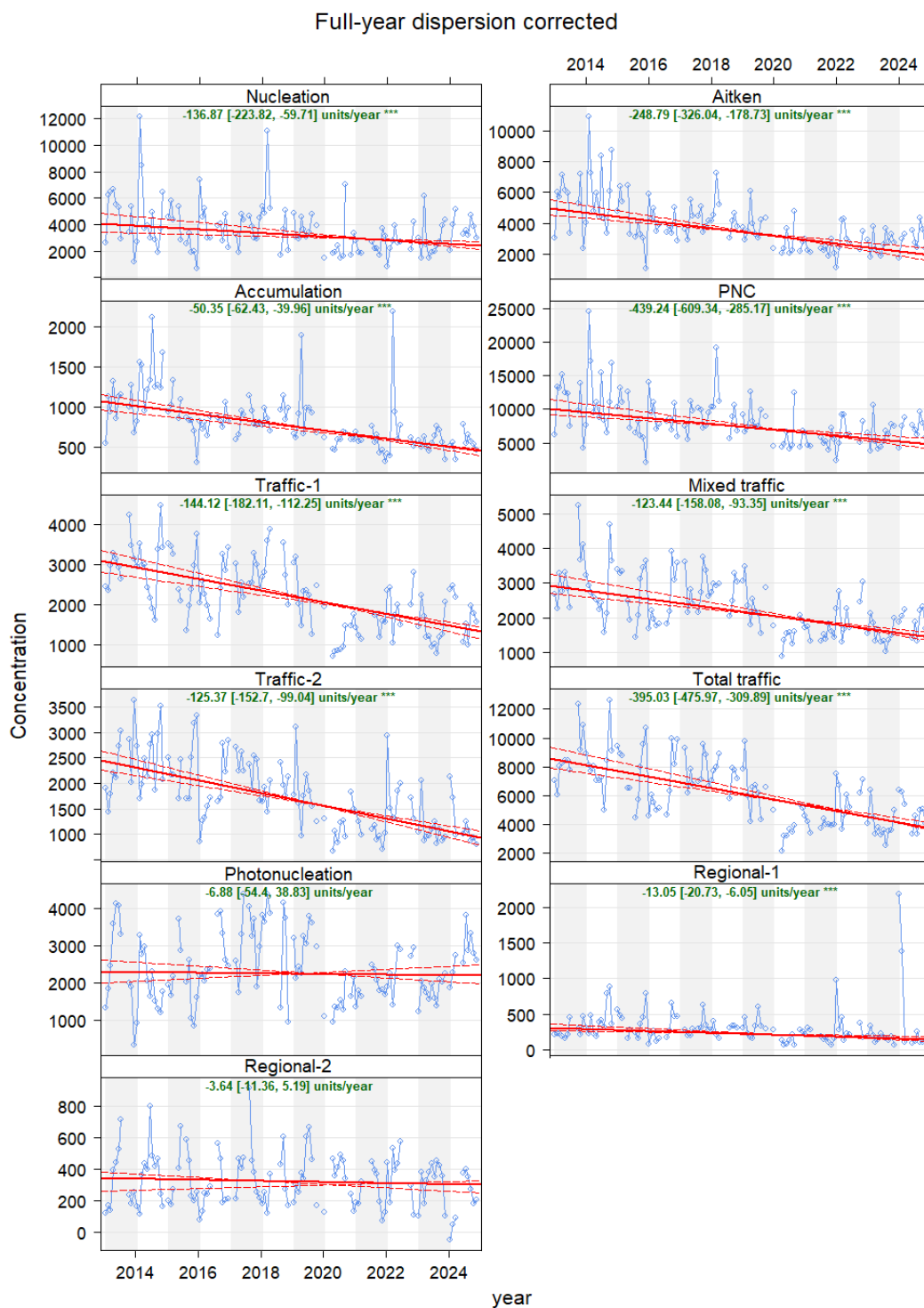


Figure 5. Trend analysis of full-year, dispersion corrected particle number concentration (PNC), size modes (Nucleation, Aitken, and Accumulation), and source contributions to PNC (Traffic-1, Mixed traffic, Traffic-2, Total traffic, Photonucleation, Regional-1, and Regional-2) (all in $\# \text{ cm}^{-3}$). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “***” ($p<0.001$), “**” ($p<0.01$), “*” ($p<0.05$), and “+” ($p<0.1$).

Photonucleation exhibited a decreasing but non-ss trend, which may be related to the variable influence of NPF events and SO₂ emissions (mostly shipping in the study area). In particular, the lowest contributions occurred in 2020–2021, largely reflecting the absence of cruise ship activity during the COVID-19 period, when no cruises operated in 2020 and only a limited number returned in 2021, while bulk shipping did not decrease markedly in this period (Figure S18; Port of Barcelona, 2025). In contrast, *Nucleation growth* (for the warm period) showed a ss decreasing trend. This reduction may reflect the combined effect of lower concentrations of condensable vapours and reduced CS associated with declining PNCs. Furthermore, the decrease of the S content in shipping fuels since 2020 may have reduced the concentrations of newly formed particles that grew while reaching the city.

Regional-1 (higher in winter) also exhibited ss significant decreases, most likely associated with reductions in primary PM emissions and precursor gases of UFP. In contrast, *Regional-2* (higher in summer) did not show a ss decreasing trend. Despite the observed reductions in SO₂ emissions, the relatively high NH₃ concentrations in the region (Liu et al., 2024), and probably the weak VOCs decreases, together with the lack of decrease in O₃ (Massagué et al., 2023), likely prevented a more marked decrease in this contribution compared with *Regional-1*.

4. Conclusions

This study provides a comprehensive evaluation of ultrafine particle (UFP) sources in Barcelona (NE Spain) over a 12-year period, assessing the influence of meteorology-driven dispersion on particle number concentrations (PNCs), as well as seasonal variability and temporal trends. Dispersion normalisation using the ventilation coefficient (VC), calculated from boundary layer height (BLH) and the wind speed (WS), was applied to an hourly particle number size distribution (PNSD) dataset from 2013–2024. The dataset was divided into two subsets: the cold season (October–March) and the warm season (April–September).

Source apportionment (SA) by positive matrix factorisation (PMF) was performed on both dispersion corrected (DC) and uncorrected (UC) datasets for each season. Seven distinct factors contributing to PNCs were identified: *Traffic-1* (mostly gasoline), *Traffic-2* (mostly diesel), *Mixed traffic*, *Photonucleation* and *Nucleation growth* (both highly influenced by shipping), *Regional-1* (higher in winter), and *Regional-2* (higher in summer). Total road traffic was the largest contributor to PNCs, accounting for 60% and 53% of the average PNC during the cold and warm seasons, respectively, according to the DC-SA. In the UC-SA, traffic contributions were higher, accounting for 69% and 58% during the cold and warm seasons, respectively. Because traffic emissions are primarily local and strongly influenced by dispersion conditions, the lack of correction for dispersion can lead to

overestimation of their contributions, as reflected in the higher UC-SA values. In contrast, the DC-SA accounts for these dispersion effects, providing source contributions that more accurately represent emission strength rather than meteorological variability.

Photonucleation (combined with *Nucleation growth* during the warm season) showed a marked increase in its contributions in the DC-SA, accounting for 34% and 39% of PNCs during the cold and warm seasons, respectively, compared to 26% and 35% in the UC-SA. This increase is consistent with the fact that photochemical nucleation typically occurs around midday, when dispersion conditions are more favourable due to enhanced atmospheric mixing.

Trend analysis revealed consistent decreases in total PNCs, size modes (nucleation, Aitken, and Accumulation), and traffic-related source contributions, with similar trends in the DC and UC datasets. Total PNCs declined by $-4.4\% \text{ yr}^{-1}$, with the strongest reduction in the Aitken mode, likely due to decreased road-traffic emissions following EURO 5 and EURO 6 regulations, the introduction of diesel particles filters (DPFs), and the implementation of the low emission zone (LZE) in Barcelona. Seasonal analyses showed more pronounced decreases during the cold season, coinciding with higher traffic intensity. Traffic-related sources declined by $-4.6\% \text{ yr}^{-1}$, with the largest decrease in *Traffic-2*, which is mainly associated with diesel emissions. The lowest traffic contributions were observed during 2020–2021, reflecting the sharp reduction in mobility during the COVID-19 lockdowns, with levels increasing again afterwards but remaining below pre-pandemic values.

Photonucleation showed a non-significant decrease, likely due to the variable influence of new particle formation (NPF) events and contributions from different sources such as shipping emissions. In this case, *Photonucleation* contributions were also strongly reduced in 2020–2021 due to the absence of cruise activity in the Port of Barcelona during the COVID-19 restrictions, followed by a rebound in 2022 as cruise traffic resumed. However, this rebound did not reach pre-2019 concentrations, most probably due to the implementation of the IMO 2020 standard, reducing sulphur (S) content in shipping fuels from 3.5% to 0.5% since January 2020. *Nucleation growth* and *Regional-1* exhibited steady declines, while *Regional-2* showed no clear trend, consistent with limited reductions in NH_3 , VOCs, and O_3 .

Overall, these results demonstrate the importance of accounting for meteorological dispersion when assessing source contributions to PNCs and highlight the effectiveness of emission control policies in reducing traffic-related UFPs. However, it should be noted that for health studies it is very important to use the UC-SA, because this reflects the true exposure.

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5 DISCUSSION

The results obtained in this thesis offer a comprehensive assessment of the spatial and temporal variability of UFPs across diverse environments (mostly urban) in Europe, with particular emphasis on identifying and quantifying the sources contributing to PNC.

This chapter discusses the main findings of the study, examining UFP characteristics and source contributions across different site types and emission contexts (Section 5.1), inter-annual trends in PNCs (Section 5.2), methodological considerations for SA (Section 5.3), and the implications of these results for AQ management and policy (Section 5.4).

5.1 Ultrafine particle characteristics and sources

5.1.1 Urban patterns across Europe

Urban environments across Europe provide essential contexts for assessing UFP exposure, as they combine dense populations with the influence of multiple emission sources. This thesis covers a broad set of monitoring sites (eighteen UB, six TR, four SUB, and one RB), offering a comprehensive basis for examining both the temporal and spatial variability of UFP concentrations and their underlying sources, which is crucial for evaluating population ambient air exposure and developing effective mitigation strategies. **Paper I** examined the levels, spatiotemporal variability, and size distribution of UFPs across these sites, while **Paper III** focused on identifying and quantifying the sources driving these patterns.

To ensure robust comparisons across the diverse monitoring site, PNSDs were harmonised, accounting for the differences in instrument detection limits and data quality issues, as described in the methodology. Figure 5.1 illustrates the impact of data harmonisation. The left panel shows the original data, including measurement artefacts in the smallest size bins and values outside the 10–800 nm size range, while the right panel presents the harmonised PNSDs used in subsequent analyses. Prior to harmonisation, the minimum detection sizes across the sites ranged from 3 to 20 nm, which substantially

affect concentrations in the nucleation mode. For example, at HEL_UB, limiting the measured range from 3–794 nm to 10–794 reduced total PNC by 14% and the nucleation mode concentration by 33%. This shows how raw measurements could misrepresent spatial variability if lower detection limits significantly differ across sites. Harmonisation removed these artefacts, standardised the size range, and ensured that inter-site comparisons accurately reflect genuine differences in PNCs. This is not essential only for scientific consistency but also to support the development of effective AQ legislation and robust epidemiological studies, as accurate and comparable measurements form the basis for assessing population exposure and potential health impacts.

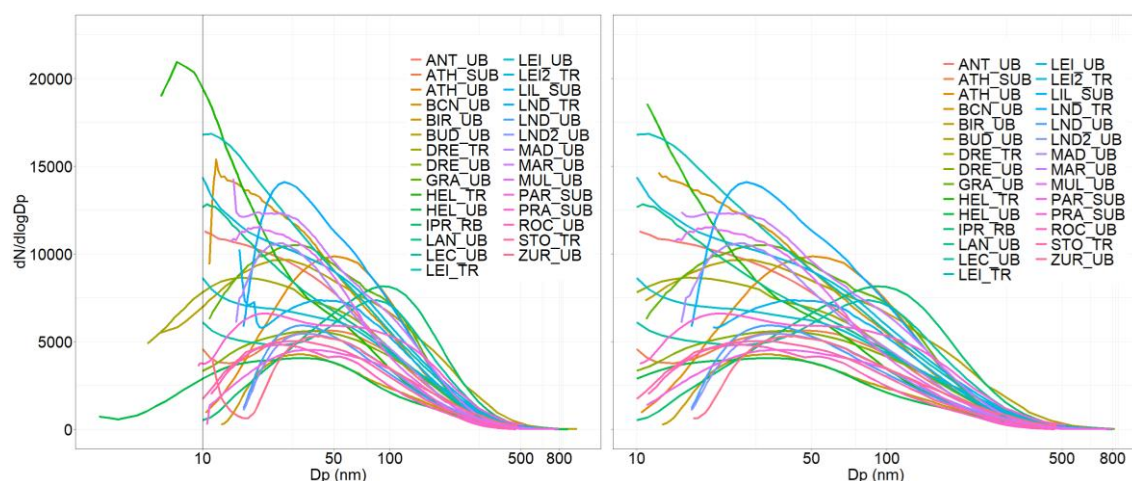


Figure 5.1: PNSDs across all monitoring sites included in **Papers I, II, III, and V** for original data (left) and harmonised data (right) after removing measurement artefacts and standardising size ranges to 10–800 nm.

Across Europe, the harmonised monitoring datasets revealed marked differences in PNCs and PNSDs (Table 5.1), reflecting the diversity of emission environments as well as climate and atmospheric conditions. TR sites consistently exhibited the highest PNCs, with an average of $11,194 \text{ # cm}^{-3}$ during 2009–2019, whereas UB and SUB/RB sites showed lower averages ($8,086$ and $6,261 \text{ # cm}^{-3}$, respectively). Nevertheless, the wide range observed at UB sites, spanning from $3,532$ to $11,318 \text{ # cm}^{-3}$, can be attributed to pronounced regional variations (see below). In some cases, PNCs at UB sites reach levels

comparable to those typically measured at TR sites in other European cities. If the UB–TR comparison is restricted to a single region (i.e., Scandinavia), the difference in PNCs is typically a factor of 2.0 to 3.0 between UB and TR sites. Regarding PNSDs, the Aitken mode dominated PNCs across all three site types, although nucleation mode concentrations were also substantial at TR sites and in some UB sites. It should be noted, however, that nucleation mode values were strongly influenced by differences in the lower detection limits of the instruments deployed at each site, as described in the methodology.

Table 5.1: Summary of PNC (in # cm⁻³) and size modes (nucleation, Aitken, and accumulation) across urban Europe, grouped by site type (UB, TR, SUB/RB), for the period 2009–2019 (where data were available). Values are expressed as mean ± standard deviation (minimum–maximum).

Site type	Nucleation	Aitken	Accumulation	PNC
UB	2,336 ± 1,346 (344–4,628)	4,232 ± 1,220 (2,197–5,894)	1,431 ± 408 (673–2,255)	8,086 ± 2,570 (3,532–11,318)
TR	4,306 ± 1,459 (2,328–5,573)	5,277 ± 1,917 (3,198–8,816)	1,723 ± 773 (851–3,045)	11,194 ± 2,359 (7,342–13,849)
SUB/RB	1,199 ± 657 (560–2,314)	3,484 ± 676 (2,517–4,148)	1,539 ± 610 (878–2,543)	6,261 ± 1,242 (4,427–7,707)

The average PNCs obtained in this study are consistent with those reported in other investigations of European cities. The mean PNC for UB sites lies below the 10,000 # cm⁻³ threshold suggested by Cassee et al. (2019), and the concentration ranges are similar to those reported in other European cities in individual studies (Keuken et al., 2015; Hofman et al., 2016; Rivas et al., 2019; Sun et al., 2019). Ridolfo et al. (2024a) reported a global average of 26,000 # cm⁻³ for TR sites, although most of the studies included in that review had size detection limits below the 10 nm cut-off used in this thesis. For European countries, average TR site values ranged from 8,400 to 25,000 # cm⁻³ (Morelli et al., 2015; Vouitsis et al., 2015; Wang et al., 2016; Sun et al., 2019; Chatain et al., 2021), reflecting differences in study period, traffic intensity, and size detection limits.

Differences between sites are not solely attributable to site type, but also reflect broader spatial patterns across Europe, as described in **Paper I**. For TR sites, spatial trends could

not be evaluated, since the data available for this thesis were limited to Northern and Central Europe and the United Kingdom (UK) and did not include Southern or Eastern Europe. By contrast, UB sites covered a wider geographical range, enabling a more detailed analysis of regional differences. PNCs exhibited a clear gradient, with lower values in Northern and Central Europe (minimum of $3,500 \text{ \# cm}^{-3}$) and higher concentrations in Southern and Eastern Europe (maximum of $11,300 \text{ \# cm}^{-3}$). A similar north–south/east trend was observed for the Aitken mode and for BC, indicating higher pollution levels in southern and eastern regions, largely associated with road traffic, as road traffic is the dominant source of BC in European urban areas (Reche et al., 2011). Nevertheless, other sources such as domestic and commercial heating can also contribute to both Aitken mode particles and BC concentrations (Casquero-Vera et al., 2021).

The spatial pattern for the nucleation mode differed slightly from those of PNC and the Aitken mode, with elevated concentrations in Southern Europe (up to $4,000 \text{ \# cm}^{-3}$) but also in central Europe (up to $4,600 \text{ \# cm}^{-3}$), primarily driven by NPF. In southern regions, high concentrations were associated with photochemical NPF, enhanced by strong solar radiation (Kulmala and Kerminen, 2008), as well as emission of gaseous precursors from shipping activities (Petzold et al., 2008; Ridolfo et al., 2025). In central Europe, plumes from SO_2 -emitting coal- or fuel-oil fired power plants also contributed to elevated nucleation mode concentrations (Junkermann et al., 2016). Fumigation processes, whereby the growth of the BLH entrains particle-air from high altitudes enriched by NPF, may affect surface concentrations. It should be noted, however, that differences in the nucleation mode across sites are partly attributable to variations in the lower detection limits of the instruments used.

These spatial trends highlight the combined influence of regional atmospheric conditions and local emission sources on UFP levels. To attribute these patterns to their underlying causes, **Paper III** applied SA across largely the same set of monitoring sites and identified ten factors contributing to UFPs. Road traffic was the dominant source, represented by four distinct traffic-related factors (*Traffic-1*, *Traffic-2*, *Mixed traffic*, and *Traffic nucleation*), alongside *Photonucleation* and *Domestic heating* at some sites. Additional contributions arose from *Urban background*, two regional sources (*Regional-1* and

Regional-2), and *Long-distance transport*. Their relative contributions varied across Europe, reflecting both local emission characteristics and regional atmospheric processes.

Although road traffic was the main contributor at nearly all sites, the SA results from **Paper III** revealed distinct secondary influences that help explain the temporal behaviours observed in **Paper I**. Based on these additional contributions, sites can be classified into three broad groups, each consistent with characteristic temporal patterns. Figure 5.2 illustrates one representative site per group, combining source contributions with daily and seasonal patterns of PNC and BC:

- High photonucleation influence: At these sites, photonucleation emerged a major secondary contributor in addition to traffic. This source profile corresponds to the temporal behaviours described in **Paper I** as “midday/morning PNC dominated sites” and, in some cases, “intermediate sites with prevalence of midday peaks”. These sites typically exhibited a strong midday peak compared with the traffic rush hours, along with higher PNCs in summer (followed by spring and autumn). Temporal patterns were anti-correlated with BC, which peaked in winter and during traffic rush hours. The observed midday enhancements reflect photochemically induced NPF events, strengthened by high solar radiation, fumigation from higher atmospheric layers enriched with nucleation mode particles, and/or contributions from nucleated particles associated with shipping, aviation, power plants and industrial emissions (Kulmala and Kerminen, 2008; Petzold et al., 2008; Junkermann et al., 2011, 2016; Rivas et al., 2019; Salma et al., 2021; Stacey et al., 2020, among others). An illustrative example is LEI_UB (Figure 5.2), where photonucleation accounted for a substantial fraction of PNCs and midday peaks were clearly distinguishable. Other sites showing similar behaviours include ANT_UB, BCN_UB, BUD_UB, DRE_UB, LEI_TR, ATH_SUB, and PRA_SUB.
- High domestic heating influence: At these sites, domestic heating was the most important secondary contributor alongside traffic. This source profile is consistent with temporal behaviours observed in **Paper I** at certain sites, where elevated PNCs were recorded during winter and at night, coinciding with peak BC concentrations. These patterns reflect the influence of residential combustion processes, particularly wood and coal burning, including cooking and heating activities from wood-burning

and pellet stoves, fireplaces, electric heaters, and kitchen stoves, among others (Tiwari et al., 2014; Zhang et al., 2014; de la Sota et al., 2018; Wang et al., 2020; Kuye and Kumar, 2023, among others). An illustrative example is LEC_UB (Figure 5.2), where domestic heating contributed substantially to PNC levels and pronounced night-time and winter peaks were evident. Other sites with similar behaviours include ATH_UB, ZUR_UB, and IPR_RB (with high contributions of domestic heating also quantified at PRA_SUB).

- Traffic-dominated with minimal other influences: At these sites, traffic was by far the dominant contributor to PNCs, while secondary sources such as photonucleation or domestic heating played only a minor role. This group corresponds closely to the “road traffic dominated sites” described in **Paper I**, as well as some “intermediate sites with greater prevalence of traffic”. Their temporal profiles were characterised by pronounced rush hours peaks in both PNC and BC, and in some cases lower concentrations during summer, reflecting the strong influence of vehicular emissions (Ogulei et al., 2007; Rodríguez and Cuevas, 2007; Harrison et al., 2011; Liu et al., 2014; Rivas et al., 2019; Sun et al., 2019; Hopke et al., 2022, among others). An illustrative example is MAD_UB (Figure 5.2), where traffic accounted for most of the PNC and the daily cycle was tightly coupled to road activity. Other sites with similar behaviours include GRA_UB, HEL_UB, LND_UB, MUL_UB, DRE_TR, HEL_TR, LND_TR, STO_TR, LIL_SUB, and PAR_SUB.

Taken together, the integration of spatial variability (**Paper I**) with SA (**Paper III**) provides a comprehensive overview of the drivers of UFP concentrations across European urban environments. Road traffic emerged as the dominant contributor at nearly all sites, but secondary influences from photonucleation and domestic heating explained much of the variability in temporal and spatial patterns.

It should be noted, however, that certain specific sources are not fully captured within this general framework. Aviation, for instance, was not resolved as a distinct source in any of the sites included in **Paper III**, yet indications of its contribution were observed at particular locations. Because of its growing relevance for urban exposure, the following section addresses aviation and other specific environments in more detail, complementing the broader European patterns discussed above.

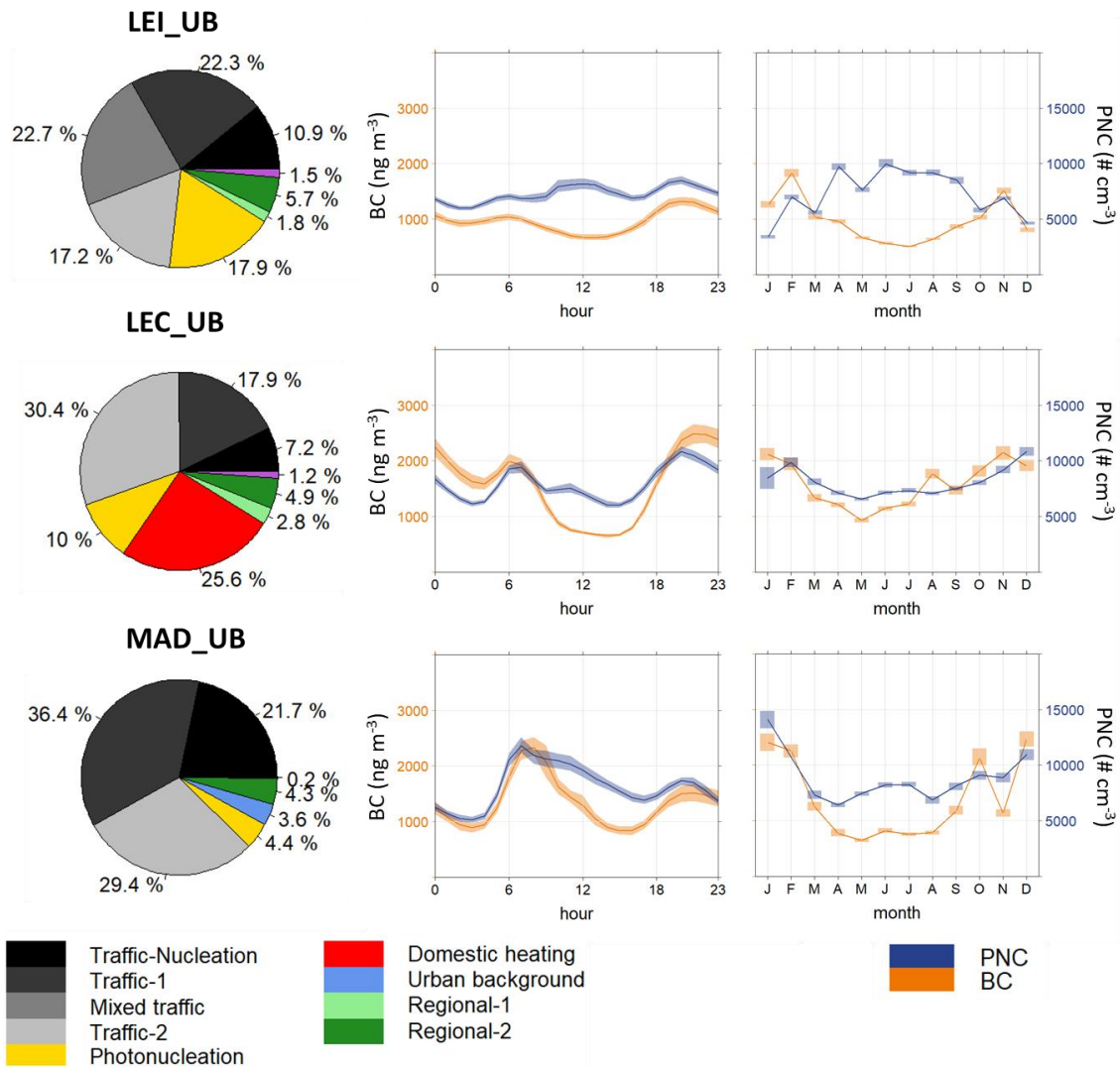


Figure 5.2: Source contributions to PNC (left, from **Paper III**) and the corresponding daily and seasonal patterns of PNC and BC (middle and right, from **Paper I**) at three representative urban sites for each source-based group: LEI_UB (high photonucleation influence, top), LEC_UB (high domestic heating influence, middle), and MAD_UB (traffic-dominated with minimal other influences, bottom). Purple in the source contributions indicates the fraction of PNC not assigned to a specific source.

5.1.2 Source influences in specific environments

The general urban patterns and dominant sources described above do not fully capture the complexity of certain environments. Airports, in particular, represent highly specific settings where emissions from aircraft operations and associated activities can exert a substantial influence on PNCs. Examining such environments is therefore necessary to complement the broader European overview and to better understand the sources affecting UFP levels. Insights from a dedicated case study at a major European airport (**Paper IV**), together with additional analyses for UB and SUB sites from **Paper III**, provide a more complete perspective on the role of aviation in shaping UFP exposures.

Paper IV offered a detailed investigation of UFP concentrations near Brussels Airport (Belgium), designed to quantify the impact on aircraft operations on ambient AQ. This study was included in the thesis because of the availability of high-quality measurement data from a two-month campaign in 2015 with a well-structured monitoring design. The four sites distributed along the runway axis (downwind and upwind) enabled SA to robustly characterise airport-related contributions not only near the runway but also up to 7 km downwind. This is particularly relevant in the context of urban AQ, demonstrating that airports can still exert an influence on PNCs at distances relevant for surrounding populations.

The results demonstrated that aircraft operations significantly affected ambient UFP levels, particularly in the nucleation mode (10–30 nm range in this study), which accounted for approximately two-thirds of the total PNC near (250–750 m) the airport (Table 5.2). Closer to the runway, average PNCs were 17,000–18,500 # cm⁻³, with nucleation mode concentrations of around 11,000 # cm⁻³. Although concentrations declined with distance, an airport-related signal remained discernible 7 km downwind, where average PNCs and nucleation mode concentrations were about 7,700 and 4,000 # cm⁻³, respectively. At the UB site A, located upwind of the airport, the average PNC (around 10,000 # cm⁻³) was consistent with typical long-term UB levels (Hofman et al., 2016) but remained higher than the average derived from the European UB sites reported in **Papers I** and **III** (around 8,000 # cm⁻³, see above).

Table 5.2: Average total and nucleation mode PNC at the four monitoring sites in **Paper IV**, with corresponding site-airport distances.

Site (type)	Site-airport distance	PNC (# cm ⁻³)	Nucleation (# cm ⁻³)
A (UB)	5 km	10,000	4,800
B (UB)	250 m	18,500	11,000
C (UB)	750 m	17,000	11,200
D (Rural)	7 km	7,700	4,000

Aircraft activity data confirmed strong correlations between UFP concentrations and landing and takeoff (LTO) operations, but attempts to distinguish the PNSD profiles of landings versus takeoffs were inconclusive. However, van Loenen et al. (2025), at a monitoring site located about 1 km from the endpoint of the nearest runway of Boston Logan International Airport (US), were able to differentiate between landings and takeoffs. They found that hourly average PNC was nearly 4 times higher during landings than during takeoffs, with size modes peaks ranging from 7 to 18 nm during landings and around 39–52 nm during takeoffs. Nonetheless, the SA in **Paper IV** provided clear insights into airport-specific factors. Six factors were identified as contributors to PNC, with aircraft-related sources accounting for up to 64% of total PNC at sites closest to the airport and up to 34% at more distant sites. These aviation factors were characterised, among other features, by their modal diameters in the nucleation mode (size modes between 12 and 20 nm). This finding is consistent with literature, which reports aircraft emissions as major contributors to particles <20 nm in size (Kinsey et al., 2010; Mazaheri et al., 2013; Lobo et al., 2015; Masiol et al., 2016; Tremper et al., 2022; Ridolfo et al., 2024b). The predominance of emissions in the nucleation mode, however, is not exclusive to aviation and similar PNSDs may also arise from photonucleation or nucleated particles from road traffic. In urban environments, this overlap can complicate the attribution to nucleation mode particles specifically to aircraft.

For this reason, although aviation was not identified as a distinct factor in the SA of **Paper III**, an additional investigation was conducted for UB and SUB sites located near major airports to explore potential aviation-related influences. Sites situated more than 20 km from the nearest airport, or those close to airports with relatively low passenger and flight activity, were excluded, as contributions from aviation at such distances or under limited

air traffic volumes are unlikely to be substantial. TR sites were also excluded, since their high levels of local road traffic emissions would mask and confound any potential signal from aviation.

To further investigate whether aviation-related signals could be detected in the broader European dataset, polar plots of total PNC, nucleation mode particles, and nucleation-related sources (*Photonucleation* and *Traffic-Nucleation*, as identified in the SA of **Paper III**) were examined at the selected sites. This focus on the nucleation size range was motivated by the findings from **Paper IV** and previous literature, which consistently show that aircraft emissions predominantly occur in this mode, while potential aviation influences might be masked when examining total PNC. Concentrations were analysed in relation to wind direction and speed, with particular attention to the orientation of the closest airport relative to each site, allowing direct assessment of whether elevated PNCs coincided with the airport sector. Airport influence was then categorised as “Clear,” “Weak,” or “None” based on visual inspection of the polar plots. The results of this analysis are summarised in Table 5.3, while Figure 5.3 and Figure 5.4 present the complete set of polar plots for PNC and nucleation mode concentrations, respectively, across all sites.

*Table 5.3: Distance and orientation of sites relative to the nearest major airport, with observed airport-related contributions to total PNC, nucleation mode particles, and nucleation-related sources (SA Nucleation, including Photonucleation and/or Traffic-Nucleation) identified in **Paper III**. Airport influence was classified as “Clear”, “Weak”, or “None” based on polar plots.*

Site	Site-airport distance	Bearing (cardinal)	Airport influence		
			PNC	Nucleation	SA nucleation
ATH_UB	20 km	100° (E)	None	None	None
BCN_UB	10 km	200° (SSW)	Clear	Clear	Clear
BUD_UB	15 km	120° (ESE)	Weak	Weak	Clear
HEL_UB	13 km	0° (N)	Weak	Clear	Clear
LEI_UB	16 km	300° (WNW)	None	None	None
LND_UB	18 km	240° (WSW)	Clear	Clear	Weak
MAD_UB	14 km	60° (ENE)	None	None	Weak
MUL_UB	20 km	200° (SSW)	Weak	Weak	Weak
ZUR_UB	9 km	350° (N)	Weak	Weak	Weak
ATH_SUB	13 km	120° (ESE)	None	None	None
PAR_SUB	12 km	80° (E)	Clear	Clear	–
PRA_SUB	8 km	250° (WSW)	Weak	Clear	Clear

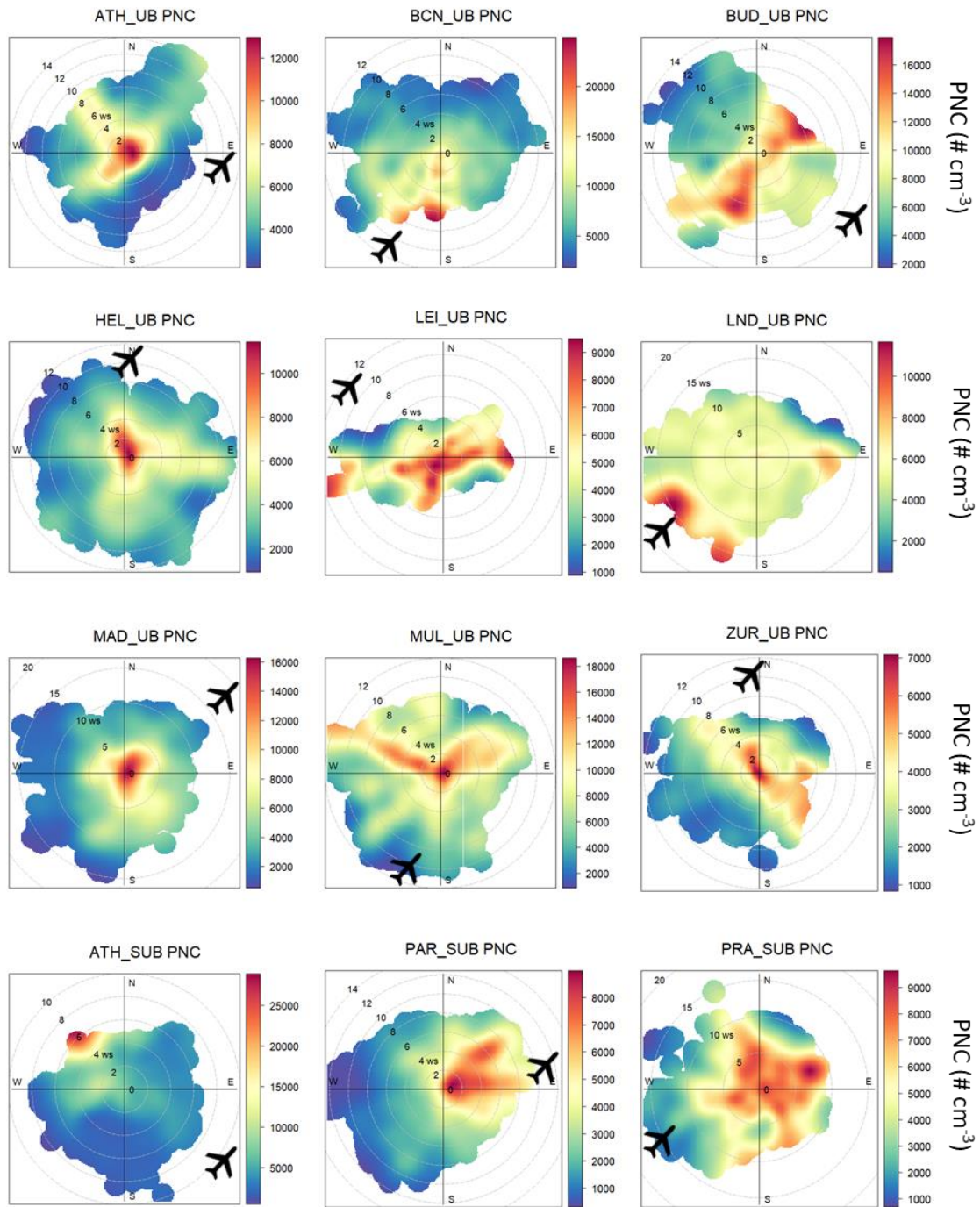


Figure 5.3: Polar plots illustrating how total PNC vary with WD and WS at 9 UB and 3 SUB sites, indicating the orientation of the nearest major airport relative to each site.

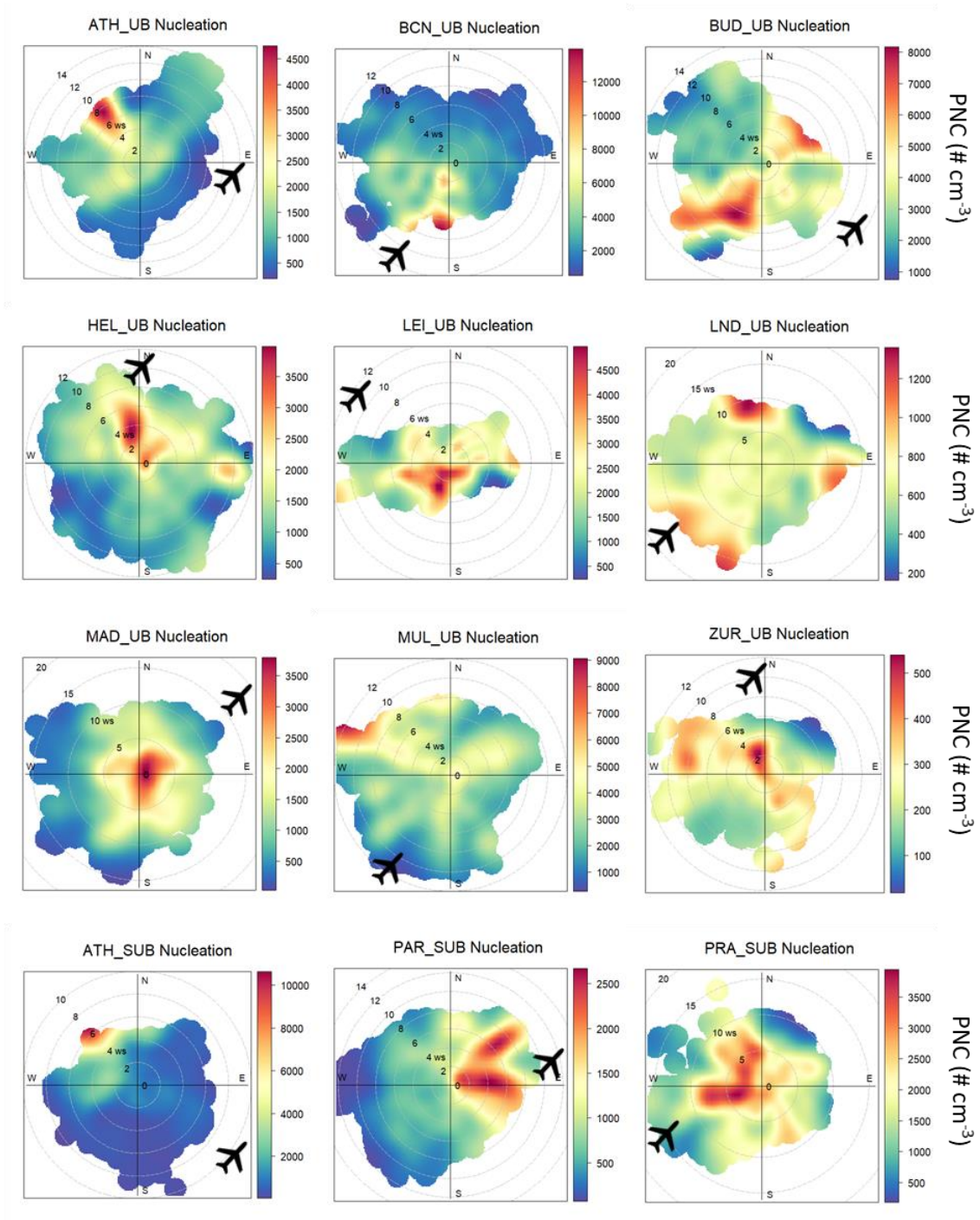


Figure 5.4: Polar plots illustrating how nucleation mode PNC vary with WD and WS at 9 UB and 3 SUB sites, indicating the orientation of the nearest major airport relative to each site.

The analysis of polar plots revealed that aviation-related contributions were detectable at several sites, but the strength of the signal varied considerably (Table 5.3 and Figure 5.3 and Figure 5.4). Clear influences were evident at BCN_UB, HEL_UB, LND_UB, PAR_SUB, and PRA_SUB, with contributions most pronounced in the nucleation mode. Weaker signals were detected at BUD_UB, MUL_UB, and ZUR_UB, suggesting greater local interference from other sources. These results support the evidence from **Paper IV** and previous literature (Cheung et al., 2011; Hudda et al., 2014; Keuken et al., 2015; Hofman et al., 2016; Masiol et al., 2017) that airport emissions can contribute to nucleation mode PNC even at distances of several kilometres from the runway.

The comparison between polar plots and sources identified in **Paper III** provided further insight into how aviation signals were represented in the factor analysis. At BCN_UB and HEL_UB, SA attributed almost all nucleation mode particles to a single source (*Photonucleation* at BCN_UB and *Traffic-Nucleation* at HEL_UB), consistent with the polar plots, which closely mirror the patterns seen for nucleation mode concentrations (Figure 5.5). This indicates that aviation-related contributions were effectively included into broader nucleation categories from the SA. In contrast, at BUD_UB, the nucleation mode particles were apportioned to both *Photonucleation* and *Traffic-Nucleation*. While the *Traffic-Nucleation* source clearly reflected airport-related influence, the *Photonucleation* source did not, resulting in a split of the nucleation mode contribution between two sources with distinct spatial patterns.

A similar approach was adopted by Rivas et al. (2019), who combined SA and polar plots at several of the same sites (BCN_UB, HEL_UB, LND_UB, and ZUR_UB). Their results also showed that aviation contributions are often embedded within broader nucleation categories in SA, consistent with the patterns observed in the present analysis.

These findings illustrate both the value and the limitations of SA in resolving aviation influences. While polar plots can reveal airport-related contributions, especially when guided by knowledge of expected size ranges, SA tends to include these contributions under more generic nucleation categories, such as photonucleation or nucleated particles from traffic. As a result, aviation emissions may remain hidden in SA results unless specifically investigated. This underscores the importance of combining receptor

modelling with directional analyses when assessing the role of airports in urban UFP exposures.

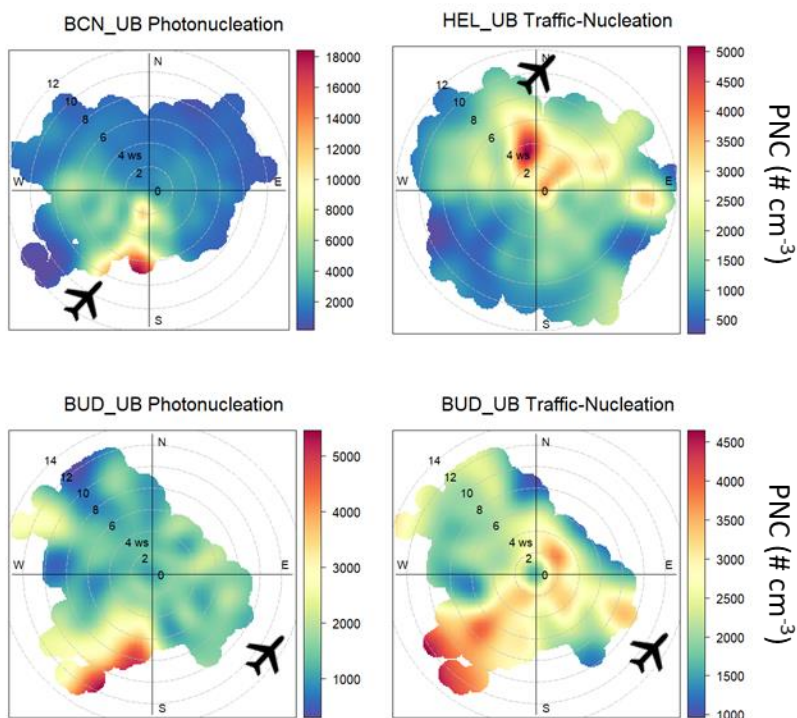


Figure 5.5: Polar plots of nucleation-related sources at BCN_UB (Photonucleation), HEL_UB (Traffic-Nucleation), and BUD_UB (Photonucleation and Traffic-Nucleation), indicating the orientation of the nearest major airport relative to each site.

Beyond aviation, other specific environment also showed clear source-related signatures in the European dataset. At the coastal site of BCN_UB, shipping contributions were evident in both SA and polar plots, pointing to high concentrations of particles in the nucleation mode from the south, where the harbour of Barcelona is located. This is consistent with previous studies highlighting the role of marine traffic in urban particle exposures (Wang et al., 2019; Merico et al., 2020; Ridolfo et al., 2025). Powerplants and industrial hotspots were also identified at several location, often characterised by distinct size modes and directional patterns. These industrial and shipping emissions may also enhance regional nucleation by releasing precursors of UFP (Li et al., 2023).

Another possible reason for the non-identification of shipping and aviation contributions in urban SA results is the growth of the nucleation mode particles as they cross suburban areas with relatively high emissions of condensable gases. Guo et al. (2019) showed that during NPF in clear air, growth rates can range from 5 to 20 nm h⁻¹. Under polluted conditions, however, pre-existing particles can suppress nucleation, thereby masking NPF events.

Taken together, these findings emphasise that airports, harbours, industrial areas, and other specific environments can exert measurable impacts on ambient primary and secondary PNCs. Importantly, such sources may affect populations beyond their immediate vicinity, as demonstrated for aviation. Thus, environments dominated by these sources require dedicated studies and tailored monitoring strategies to capture their contributions, which might otherwise be underestimated in broad urban averages. These hotspots should therefore be monitored as required by the Ambient Air Quality Directive (EU) 2024/2881, using fixed sampling points positioned to capture maximum population exposure and ensure their impacts are fully quantified (EC, 2024).

5.2 Inter-annual trends of ultrafine particles

Understanding the evolution of UFP concentrations over time is essential for evaluating the effectiveness of policy measures and suggest new ones for further abatement of pollution. **Paper II** analysed inter-annual trends in UFP concentrations and different size modes (total PNC, nucleation, Aitken, and accumulation) across multiple urban sites in Europe in 2009–2019, while **Paper III** extended this perspective by evaluating long-term trends in the contributions of the sources identified, at (mostly) the same sites and periods, by using PMF receptor modelling. Additionally, **Paper V** provided a detailed case study for BCN_UB, extending the analysis from 2013 to 2024, and applying dispersion normalisation to better separate emission-driven changes from meteorological variability.

5.2.1 Multi-site European trends

Paper II showed clear reductions in PNC during 2009–2019, but these reductions were not uniform across size modes and site types. As summarised in Table 5.4a, the meta-

analysis showed that total (mostly urban) PNC declined moderately at the European scale, largely driven by decreases in the Aitken and accumulation modes. In contrast, nucleation mode concentrations displayed no ss long-term change on average, with slight (non-ss) increases observed at UB sites.

A major driver of the observed decreases was the tightening of European vehicle emission standards over the past two decades. The EURO emission standards, first introduced by the EU in 1992, were designed to reduce the environmental and health impacts of pollutants emitted by vehicles (EC, 1991). These regulations have progressively become stricter. Starting with the EURO 5 standard (implemented in 2011), a particle number (PN) limit per kilometre was introduced for new light-duty vehicles (EC, 2007), and this requirement was maintained and reinforced in 2015 with the EURO 6 standard (EC, 2018). Equivalent EURO V and VI standards were also implemented for heavy-duty vehicles. To comply with the PN and PM emission limits, diesel particulate filters (DPFs) became mandatory.

Diesel vehicles account for most of the exhaust PM from road traffic. Diesel exhaust particles are largely solid graphitic particles (elemental carbon), with a major mode at 40–80 nm, together with smaller liquid particles formed largely from condensed engine oil vapour (Kittelson et al., 2006; Harrison et al., 2018; Hopke et al., 2022). The smallest particles form in the atmosphere by nucleation of SVOCs as emissions are diluted and cooled (Charron and Harrison, 2003).

DPFs are reported to be highly effective at removing PN and PM but they are not designed to remove vapour-phase material. As a result, the widespread adoption of DPFs led to pronounced reductions in particles emitted from vehicles, particularly in the Aitken and accumulation modes. However, as the formation of the nucleation mode particles occurs in the diluting exhaust gases, EURO emission standards were defined in terms of PNCs of non-volatile (solid) particles > 23 nm in the exhaust (EC, 2018). This, combined with contributions from other sources of nucleation mode particles such as photochemical nucleation, industry, shipping, and aircraft emissions, helps explain why nucleation concentrations did not decline significantly over the study period. Furthermore, the reduction in CS potential caused by decreases in road traffic emissions may have

facilitated NPF, contributing to increases in the nucleation mode (Kerminen & Kulmala, 2002; Korhonen et al., 2013).

The strong role of road traffic is also evident in the steeper PNC declining slope observed at TR sites compared with UB ones (Table 5.4a), reflecting the direct influence of emissions controls at sites dominated by vehicle exhaust. In contrast, UB sites are also affected by a wider mix of sources, including others of regional and local emissions, which dilute the relative effect of traffic measures. As a result, improvements in UFP levels at UB sites have been more modest, underscoring the importance of non-traffic sources in shaping overall urban exposures.

*Table 5.4: Average annual percentage changes [95% confidence intervals] (% yr⁻¹) in PNC across UB, TR, and SUB sites, as well as the global average, for: a) total PNC and nucleation, Aitken, and accumulation modes; b) traffic-related sources (Traffic-1, Traffic-2, Mixed traffic, and Traffic-Nucleation); and c) other sources (Photonucleation, Regional-1, and Regional-2). Values are derived from a meta-analysis of multi-site trends reported in **Papers II** and **III** for the period 2009–2019. Asterisks (*) indicate ss trends.*

a)

Type	Nucleation	Aitken	Accumulation	PNC
UB	1.09 [-3.27; 5.45]	-2.40* [-3.84; -0.96]	-3.69* [-4.98; -2.39]	-1.89* [-3.70; -0.09]
TR	-3.85 [-7.77; 0.07]	-5.79* [-8.11; -3.48]	-5.79* [-6.83; -4.75]	-5.13* [-7.71; -2.56]
SUB	-1.68 [-4.16; 0.81]	0.93 [-2.71; 4.57]	-0.99 [-3.13; 1.15]	0.06 [-2.04; 2.16]
Global	-0.60 [-3.20; 2.01]	-2.81* [-4.26; -1.37]	-3.92* [-4.99; -2.86]	-2.46* [-3.89; -1.04]

b)

Type	Traffic-1	Traffic-2	Mixed traffic	Traffic-Nucleation
UB	-1.81 [-3.68; 0.06]	-3.21* [-4.81; -1.60]	-2.16* [-4.11; -0.22]	-0.52 [-4.51; 3.47]
TR	-4.93* [-8.47; -1.40]	-5.93* [-7.91; -3.96]	-9.42* [-14.41; -3.76]	-4.55* [-7.99; -1.11]
SUB	3.26 [-2.47; 10.94]	-2.66* [-5.22; -0.11]	-1.68 [-3.90; 0.90]	—
Global	-2.78* [-4.78; -0.78]	-4.21* [-5.60; -2.83]	-2.97* [-5.77; -0.16]	-2.97* [-5.77; -0.16]

c)

Type	Photonucleation	Regional-1	Regional-2
UB	2.27 [-1.93; 6.47]	-5.90* [-8.90; -2.90]	-1.83 [-4.29; 0.62]
TR	4.06 [-6.39; 14.51]	-5.17* [-8.16; -2.18]	-1.86* [-3.71; -0.02]
SUB	-2.37 [-5.46; 0.72]	-1.96 [-6.62; 3.97]	-1.98 [-5.95; 2.72]
Global	1.48 [-1.70; 4.65]	-5.40* [-7.36; -3.43]	-1.90* [-3.56; -0.25]

While **Paper II** highlighted overall declines in UFP concentrations and size modes (except for the nucleation mode), **Paper III** allowed identification of the sources responsible for these trends. Table 5.4b summarises the inter-annual changes in traffic-related factors contributing to PNC during 2009–2019. This analysis confirmed that traffic-related sources were the dominant drivers of the observed decreases, particularly at TR sites where direct vehicular emissions are most influential. The strongest significant reduction was observed for Traffic-2, primarily linked to diesel exhaust emissions, consistent with the downward trends of the Aitken and accumulation modes, and reflecting the widespread adoption of DPFs. The weakest decreases were associated with Traffic-Nucleation, which is largely composed of freshly nucleated particles from SVOCs emitted by diesel engines. This supports the limited impact of DPFs on the nucleation mode (Chen et al., 2022b; Damayanti et al., 2023). Similar reductions were found for Traffic-1, including nucleation particles from diesel but also particles emitted from gasoline engines, which are reported to emit fewer particles (Rönkkö et al., 2023). The pronounced declines in Mixed traffic, especially at TR sites, further underscore the effectiveness of emission controls across diverse traffic fleets.

Beyond traffic-related sources, Paper III also examined long-term trends in secondary and regional contributors to PNC (Table 5.4c). The decrease of Regional-1, linked with nitrate, formed through atmospheric oxidation of NO_x mainly during winter and at night, might be associated with the decrease UFP NO_x precursors (EEA, 2023). A smaller reduction was also observed for Regional-2, associated with secondary ammonium sulphate and organic aerosols in summer. This source maximises with high O₃, which has increased in urban environments preventing a more marked decrease of this source contribution in spite of decreases in SO₂ emissions (Sicard et al., 2021). In contrast, Photonucleation did not exhibit significant declines and instead showed non-ss slight increasing trends at UB and TR sites. This stability or weak increase in nucleation-related source contributions is in line with the absence of long-term reductions in nucleation mode concentrations (Paper II) and may partly reflect the reduced CS from lower traffic emissions, favouring NPF events

5.2.2 Extended case study: Barcelona

To complement the multi-site analyses of **Papers II** and **III**, **Paper V** focused on Barcelona (BCN_UB) as an extended case study, providing a long-term perspective from 2013–2024. Although dispersion normalisation was applied in **Paper V** to enhance trend robustness, the results presented here are based on uncorrected data, allowing direct comparison with the prior multi-site analyses. The methodological implications of dispersion normalisation are discussed in Section 5.3. Barcelona is particularly suitable for this analysis due to its long and high-quality UFP dataset (12 years from 2013 to 2024), the numerous (EU, national, regional, and local) policy actions implemented, and its location in a southern European environment where emissions and meteorology interact strongly.

The annual mean PNC at BCN_UB from 2013–2024 is presented in Figure 5.6, with key regulatory interventions and external events annotated on the time axis. The numerical trends for size-resolved modes and source contributions are summarised in Table 5.5, allowing a detailed comparison of nucleation, Aitken, and accumulation modes, as well as traffic-related and other sources. The trend analysis revealed consistent decreases across most variables, with the strongest reductions in the Aitken and accumulation modes, while nucleation mode concentrations declined more modestly. In terms of source contributions, traffic-related factors showed the most pronounced reductions, particularly those associated with diesel emissions (Traffic-2), while non-traffic sources exhibited more heterogeneous patterns. Photonucleation and Regional-2 did not display ss trends, while Regional-1 declined moderately, underscoring the influence of secondary and background processes in shaping UFP levels in Barcelona.

Extending the analysis period to 2024 revealed a clear intensification of the decreasing trends compared with 2013–2019 (Table 5.5), reflecting the effect of successive regulatory interventions. The implementation of the EURO 5 (2011) and EURO 6 (2015) vehicle emissions standards progressively reduced exhaust particles emissions, particularly from diesel vehicles, leading to pronounced declines in the Aitken and accumulation modes and in the Traffic-2 factor, consistent with the widespread adoption of DPFs.

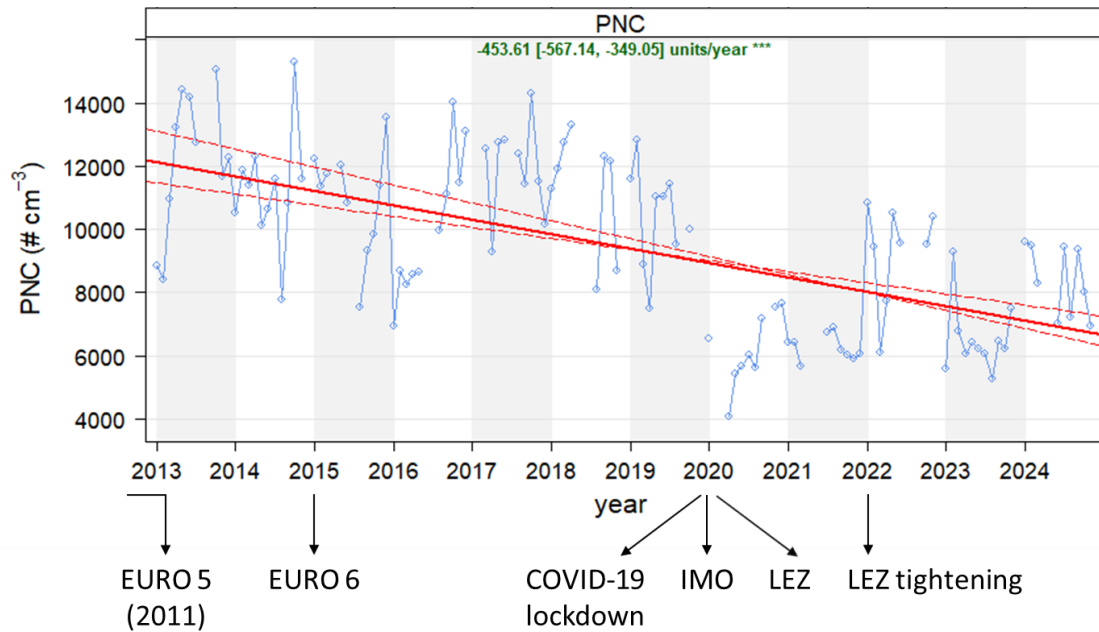


Figure 5.6: Long-term trends in annual mean PNC at BCN_UB from 2013–2024 (**Paper V**). Values are given as slopes [95% confidence intervals] in $\# \text{ cm}^{-3} \text{ yr}^{-1}$ and ss is marked by *** ($p < 0.001$). Key interventions are annotated on the time axis.

From 2020 onward, additional measures were implemented, which further strengthened the downward trend and contributed to the sharper decreases observed in the 2013–2024 period compared with 2013–2019. During 2020–2021, the COVID-19 main and relaxed lockdown measures produced a marked drop in PNCs, affecting not only traffic-related sources but also the Photonucleation factor, reflecting the absence of cruise ship activity and reduced aviation during this period (the bulk shipping was practically unaffected, Port de Barcelona, 2025). In addition, the International Maritime Organization (IMO) regulation introduced from January 2020 mandated a reduction in the sulphur content of marine fuels from 3.5% to 0.5% (IMO, 2020), leading to lower emissions of SO_2 , a key precursor of NPF. This measure likely contributed to the observed decreases in the nucleation mode and in regional source contributions influenced by shipping.

The establishment of Barcelona’s Low Emission Zone (LEZ) in January 2020 also accelerated the decline of all traffic-related factors and total PNC. After the pandemic, PNC levels increased again but remained below pre-pandemic values, partly due to the tightening of the LEZ from July 2022, which sustained the reductions in traffic-related

contributions by restricting the circulation of older, high-emitting vehicles, with particularly strong limitations on diesel fleets (Area Metropolitana de Barcelona, 2020; Ajuntament de Barcelona, 2021).

Table 5.5: Annual percentage changes [95% confidence intervals] (% yr⁻¹) in PNC at BCN_UB from 2013–2019 (**Papers II and III**) and 2013–2024 (**Paper V**) for: a) total PNC and nucleation, Aitken, and accumulation modes; b) traffic-related sources (Traffic-1, Traffic-2, and Mixed traffic); and c) other sources (Photonucleation, Regional-1, and Regional-2). Statistical significance is indicated by *** ($p < 0.001$), ** ($p < 0.01$), and * ($p < 0.05$).

a)

Type	Nucleation	Aitken	Accumulation	PNC
2013–2019 (Paper II)	2.87 [-1.09; 6.36]	-2.74*** [-4.46; -0.92]	-1.85 [-4.59; 1.13]	-0.95 [-2.71; 1.09]
2013–2024 (Paper V)	-2.45*** [-3.52; -1.50]	-4.30*** [-4.91; -3.71]	-4.40*** [-5.19; -3.45]	-3.74*** [-4.33; -3.04]

b)

Type	Traffic-1	Traffic-2	Mixed traffic
2013–2019 (Paper II)	-4.87** [-7.46; -2.13]	-4.06*** [-6.44; -1.66]	-1.60 [-3.79; 0.89]
2013–2024 (Paper V)	-4.76*** [-5.70; -3.80]	-5.04*** [-5.77; -4.28]	-4.64*** [-5.30; -4.00]

c)

Type	Photonucleation	Regional-1	Regional-2
2013–2019 (Paper II)	6.78* [1.12; 14.45]	1.38 [-3.71; 8.11]	0.63 [-3.50; 5.33]
2013–2024 (Paper V)	-1.01 [-2.39; 0.74]	-3.94** [-5.64; -1.47]	1.24 [-1.01; 3.67]

Overall, the extended time series for Barcelona highlights the cumulative impact of successive regulatory actions on UFP concentrations, with the most substantial decreases linked to measures targeting road traffic and the absence of cruising in 2020–2021, while other sources remained less sensitive to local interventions. When compared with the broader European trends reported in **Papers II and III** for 2013–2019, Barcelona exhibits stronger decreases in the Aitken mode and traffic-related factors, reflecting the effectiveness of European, national, regional, and local policies. In contrast, the slight

increase observed in the nucleation mode and Photonucleation factor suggests a more pronounced influence of regional and meteorological processes in southern European environments, where photochemical activity is stronger and cruising very common.

5.3 Methodological considerations for source apportionment

5.3.1 Dispersion normalisation

The interpretation of SA results relies strongly on the methodological approaches adopted during data processing and receptor modelling. In the context of UFP studies, dispersion normalisation can play a key role in disentangling emission-driven changes from meteorological variability and in identifying the relative contributions of the different sources more accurately (Dai et al., 2020; Chen et al., 2022; Hopke et al., 2024; Vörösmarty et al., 2024). Dispersion normalisation was applied in **Paper V** to examine source contributions to PNC and long-term trends at the UB site of Barcelona (BCN_UB), with the aim of better isolating changes associated with emission dynamics from those driven by meteorological fluctuations. Data were also analysed considering seasonal variability, divided into a cold (October–March) and a warm (April–September) period, following the seasonal variations in PNSD profiles driven by changes in atmospheric chemistry reported by Zhou et al. (2004) and Ogulei et al. (2007).

When comparing PMF results obtained from dispersion corrected (DC) and uncorrected (UC) datasets, similar source profiles and temporal behaviours were observed, although notable differences emerged in the relative source contributions. The application of DC reduced the influence of local sources, mainly traffic, by 13% and 8% in the average PNC compared with the UC results for the cold and warm seasons, respectively. Because traffic emissions are primarily local and strongly influenced by dispersion conditions (with peak emissions during rush hours occurring under low WS and shallow BLH), the lack of correction for dispersion can lead to an overestimation of their contributions, as reflected in the higher UC values. In contrast, the DC approach accounts for these dispersion effects, providing source contributions that more accurately represent true emission dynamics rather than meteorologically modified values. Conversely, DC increased the

relative contributions of photochemically driven particles by 24% and 11% compared with the UC data for the cold and warm seasons, respectively. This increase is consistent with the fact that photochemical nucleation typically occurs around midday, when dispersion conditions are more favourable due to enhanced atmospheric mixing (high WS and deep BLH).

Previous studies have also reported notable differences in source contributions when applying dispersion normalisation, highlighting the importance of accounting for meteorological variability when interpreting PNSD data, source apportionment results, and trend analyses (Dai et al., 2020a, 2020b; Chen et al., 2022a, 2022b; Hopke et al., 2024; Vorosmarty et al., 2024).

The application of DC also influenced the interpretation of long-term trends in PNCs and source contributions, providing a more stable and accurate representation of changes in emissions over time (Chen et al., 2022b). Comparing DC and UC datasets shows that the total PNC exhibited a steeper negative trend after applying dispersion normalisation, confirming that accounting for dispersion helps to better distinguish emission-related trends from those influenced by meteorological conditions (Table 5.6). However, for Total traffic and Photonucleation, the estimated slopes were very similar between DC and UC datasets, suggesting that DC had a limited effect on the long-term trends for these sources.

*Table 5.6: Annual percentage changes [95% confidence intervals] (% yr⁻¹) in PNC and source contributions from Total traffic (sum of traffic sources) and Photonucleation at BCN_UB, obtained from DC and UC datasets for the period 2013–2024 (**Paper V**). Statistical significance is indicated by *** ($p < 0.001$).*

Dataset	PNC	Total traffic	Photonucleation
DC	-4.37*** [-5.37; -3.13]	-4.64*** [-5.12; -3.93]	-0.30 [-2.08; 1.93]
UC	-3.74*** [-4.33; -3.04]	-4.76*** [-5.30; -4.18]	-1.01 [-2.39; 0.74]

The use of dispersion normalisation should, however, be carefully considered depending on the objectives of the study. For health studies, where the aim is to assess actual

population exposure, data should not be normalised, but the exposure concentrations used. In contrast, for policy-oriented studies focused on evaluating emission changes or the effectiveness of mitigation measures, dispersion normalisation should be applied, as it isolates the emission signal and provides a clearer indication of which sources need to be reduced to achieve AQ improvements.

5.3.2 Limitations of receptor modelling

While dispersion normalisation can improve the interpretation of SA results, the performance of receptor models such as PMF remains constrained by several methodological limitations. The main limitation of PMF is its assumption that source profiles are static and independent of each other (Bhandari et al., 2022; Chen et al., 2022). Consequently, these models cannot directly account for real-time processes such as particles growth from nucleation to Aitken or ultrafine to accumulation modes, NPF, or seasonal variability in source characteristics. For instance, nucleation mode particles characterised by an initial mode diameter of approximately 13 nm may grow at a rate of 5–20 nm h⁻¹ (Guo et al., 2019). After several hours, these particles can reach mode diameter of around 23 nm, altering the PNSD patterns associated with the nucleation profile.

At several sites analysed in **Paper III**, nucleation mode particles were not properly separated by PMF into the two main sources contributing to this mode: photochemical nucleation and traffic-related nucleation. These sources were distinguished by their characteristic triple-peak diurnal pattern, resulting from the combination of midday photochemical nucleation and the morning and evening increases in traffic-related particles. To address this limitation, an additional methodology was applied to separate the nucleation factor into these two sources, using NO_x and BC as proxies for traffic emissions. Furthermore, at some other sites examined in **Paper III**, as well as the site analysed in **Paper V**, particle growth within the nucleation mode was not identified by PMF and was instead mixed with regional or traffic sources due to the similarity of their PNSD profiles.

To overcome these limitations, Beddows et al. (2025) introduced the wide-PMF approach, which restructures the input data to incorporate temporal information directly

into the model. In conventional PMF (as used in this thesis), rows of hourly PNSD data are treated as independent observations, without consideration of their temporal sequence. As a result, the model cannot capture cyclic variations in source profiles, such as those following diurnal or seasonal patterns, and instead produces static factors that represent averaged rather than time-varying sources. To address this issue, wide-PMF reorganises the input data so that factors whose PNSD vary systematically over a 24-hour cycle can be resolved. Using this approach, Beddows et al. (2025) were able to better distinguish photochemical nucleation from traffic-related nucleation, thereby addressing one of the main weaknesses of conventional PMF.

Although methodological advances such as wide-PMF can partially overcome the static profile limitation of conventional receptor modelling, other modelling frameworks, such as Chemical Transport Models (CTMs), can also enhance the interpretation of UFP sources. CTMs are three-dimensional AQ models that can complement PMF by explicitly simulating atmospheric processes, including emissions, transport, and transformation mechanisms (Siouti et al., 2025; Patoulas et al., 2025). Ongoing studies by the Foundation for Research and Technology-Hellas (FORTH, Greece) and IDAEA-CSIC suggest that CTM-derived source contributions align more closely with DC results than with UC ones. Specifically, CTMs reproduce the lower relative contributions of traffic and the higher contributions of photochemical nucleation observed after dispersion normalisation, whereas UC SA results typically overestimate traffic and underestimate photochemical nucleation. However, discrepancies between DC-PMF and CTM-based SA still persist, mainly due to the inability of PMF to capture cyclic variations in source profiles and particle growth processes, features that CTMs represent more explicitly.

5.4 Policy implications for air quality

The findings from this thesis highlight the importance of considering both primary and secondary sources of UFPs in urban environments and provide guidance on potential regulatory and mitigation measures. This section discusses the main policy implications arising from the observed variability, SA results, and temporal trends.

The planned inclusion of UFPs in the new Ambient Air Quality Directive (EU) 2024/2881 (EC, 2024) represents a significant step forward in European AQ management. To ensure effective implementation, harmonised monitoring approaches, standardised instrumentation, and representative urban measurement sites will be essential. The establishment of such networks would facilitate the integration of UFP metrics into AQ assessments and enable a more comprehensive understanding of population exposure.

Urban traffic remains a dominant source of UFPs in most studied environments. Therefore, reducing traffic activity and renewing vehicle fleets with more sustainable technologies continue to be among the most effective strategies for decreasing UFP exposure. The widespread implementation of DPFs has significantly reduced solid particle emissions; however, these devices are not designed to capture SVOCs that can nucleate after exhaust release into the atmosphere. Consequently, although total PNC may decrease, nucleation mode particles can still represent a substantial fraction. This finding underscores the need for complementary measures that addresses both solid and volatile particle emissions from exhaust sources.

Given the ongoing renewal of urban vehicle fleets in Barcelona, reducing traffic density by promoting modal shifts towards public transport and active transport is highly recommended. This should be complemented by restrictions on the circulation of pre-EURO 5/V diesel vehicles, improved logistics for urban freight distribution, and vehicle electrification (MITERD, 2025). Evidence from the analysis of long-term trends in cities with LEZs further supports the effectiveness of such policies in reducing traffic-related PNCs.

Residential combustion also contributes substantially to wintertime UFP levels in several European cities, highlighting the need for stricter emission controls for domestic heating systems, particularly biomass and fossil-fuel boilers. A notable example is the Lombardy Region (Norther Italy), which has implemented progressive regulations for biomass combustion appliances. Under Regional Decrees 5360/2021 and 3649/2024, biomass boilers are classified from one to five stars according to emission performance, with only five-star units permitted for new installations (Lombardy Region, 2021, 2024). Financial incentives cover up to 65% of replacement costs when older systems are upgraded to compliant models. Additionally, agricultural biomass burning is restricted to areas above

1000 m a.s.l., with limits on the tonnes of biomass burned per hectare. These targeted measures demonstrate how regional policies can effectively mitigate emissions from residential combustion and could serve as models for broader implementation across Europe.

In addition to the road traffic and domestic emissions, nucleation processes represent one of the major sources of UFP in many cities. Photochemical nucleation was observed to play a particularly significant role under warm conditions. These findings suggest that climate change, through increasing temperatures and solar radiation, may further amplify NPF in the future. Consequently, AQ policies should account for the interactions between primary emissions and secondary formation processes under changing climatic conditions, emphasising the need to reduce gaseous precursor emissions (e.g., SO₂, VOCs, NH₃) that contribute to NPF.

Continued reductions in SO₂ emissions from shipping and power generation remain critical, as these emissions promote nucleation and growth of UFPs. The introduction of stricter fuel sulphur limits in maritime transport has already demonstrated substantial benefits and should be maintained and, where possible, expanded.

Airports also represent an important source of concern, particularly for smaller urban areas located near airport facilities, where aviation-related activities may constitute a significant local source of UFPs. The results highlight the importance of monitoring UFP concentrations in cities close to airports to better evaluate population exposure and the contribution of aviation-related emissions. This thesis indicates that taxiing emissions represent a relevant contributor to UFP emissions in airport environments. Consequently, measures aimed at optimising ground operations, including the reduction of taxiing times, could help mitigate these emissions. Nevertheless, further site-specific studies are recommended to better quantify these impacts and support the development of targeted mitigation strategies.

6 CONCLUSIONS

This thesis provides one of the most comprehensive assessments of ultrafine particle (UFP) concentrations and source apportionment (SA) across European urban environments to date, based on a harmonised dataset covering eighteen urban background (UB), six traffic (TR), four suburban (SUB), and one regional background (RB) sites. By combining spatial variability analysis, long-term trend evaluation, SA, and methodological approaches such as dispersion normalisation, this work advances the understanding of the factors controlling particle number concentrations (PNCs) and their temporal behaviours. It also evaluates the implications of these findings for air quality (AQ) policy and mitigation strategies, providing a robust evidence base to guide both scientific research and practical interventions aimed at reducing UFP exposure and protecting public health across Europe.

The compilation of particle number size distribution (PNSD) datasets in this study ensured a broad geographical coverage and enabled robust comparative analyses across diverse urban settings and climates. The harmonisation of PNSDs across sites within the 10–800 nm size range allowed reliable inter-site comparisons, minimised artefacts arising from different lower size detection limits, and strengthened the scientific basis for evaluating UFP exposure. This methodological advancement is essential not only for research consistency but also for supporting AQ legislation and epidemiological studies.

The main conclusions can be summarised as follows:

- There is a clear need to harmonise PNSD and PNC measurements across Europe, which will be facilitated by the inclusion of these new AQ parameters in the Ambient Air Quality Directive (EU) 2024/2881.
- Pronounced spatial gradients of PNCs were identified, with lower concentrations in Northern and Central Europe and higher concentrations in Southern and Eastern Europe. These patterns largely reflect the combined influence of regional atmospheric and climatic processes, as well as local emission sources, on UFP variability.

- Within specific regions, a clear PNC gradient was observed, following the pattern TR > UB > SUB, indicating that traffic is the dominant UFP source in urban environments. Data from TR sites were not available in Southern and Eastern Europe, which limits full regional comparability.
- Results from the SA analysis using Positive Matrix Factorisation (PMF) confirmed that road traffic (including distinct contributions from diesel, gasoline, nucleation, and mixed traffic) is the dominant contributor to UFP concentrations across urban Europe. Photochemical nucleation (linked to shipping, power generation, and other precursor sources) and domestic heating were also identified as important contributors, collectively shaping both the spatial and temporal variability of UFPs.
- A case study conducted at Brussels Airport demonstrated that aircraft-related emissions can dominate local UFP levels, with substantial contributions measurable not only at the runway but also up to 7 km downwind. This highlights their relevance for the population surrounding airports. SA revealed that aircraft-specific factors accounted for a large fraction of nucleation mode particles, although their signals are often masked or misattributed to broader nucleation categories. This underscores both the strengths and the limitations of SA in resolving aviation influences and the need to combine it with directional analyses. At more distant locations, particle growth and mixing with other sources may difficult the identification of airport-related contributions in PMF SA analyses.
- An extended analysis of urban and suburban sites across Europe confirmed that aviation influences are detectable at several locations, indicating that airports can exert measurable impacts on ambient PNCs and affect populations well beyond their immediate vicinity.
- Long-term trend analysis from 2009–2019 showed overall reductions in PNCs across European sites, primarily driven by decreases in the Aitken and accumulation modes. The tightening of European vehicle emissions standards, including the introduction of particle number (PN) limits for EURO 5/6 vehicles, and the widespread adoption of diesel particle filters (DPFs) were the main drivers of these reductions, particularly at traffic-dominated sites. Trend analysis on source contributions further indicated

that traffic-related factors, especially diesel exhaust emissions, contributed most to the observed declines.

- Nucleation mode concentrations and the photochemical nucleation factor remained largely stable or showed slight increases, reflecting several interacting processes: semi-volatile organic compounds (SVOCs) escaping from DPFs nucleate in the atmosphere immediately after emission; the reduced condensation sink (CS) associated with lower traffic emissions favours new particle formation (NPF) events; and additional nucleation mode particles are supplied by non-traffic sources such as industry, shipping, and aircraft emissions.
- An extended analysis covering 2013–2024 in Barcelona revealed stronger PNC decreases than those observed for 2013–2019, reflecting the cumulative effects of successive European, national, regional, and local policies implemented from 2020 onwards. These included the establishment and subsequent tightening of the Low Emission Zone (LEZ), which restricted circulation of older, high-emitting vehicles lacking DPFs, and the International Maritime Organization (IMO) sulphur limits, which reduced SO₂ emissions from shipping. The COVID-19 lockdowns, which temporarily decreased traffic and cruise activity, also contributed to these reductions. Collectively, these measures led to pronounced declines in traffic-related factors and even contributed to decreases in nucleation mode concentrations, reflecting the influence of reduced precursor emissions on NPF.
- The application of dispersion normalisation in the SA analysis for Barcelona (2013–2024) improved the interpretation of UFP sources by separating emission-driven changes from meteorological variability, using boundary layer height (BLH) and wind speed (WS) as key parameters. Dispersion corrected (DC) SA reduced traffic contributions while increasing that of photochemical nucleation, reflecting the influence of atmospheric mixing under favourable dispersion conditions, particularly around midday, with deep BLH and high WS. DC analysis also resulted in steeper declines in the long-term trends of total PNC, better capturing emission-related changes.
- The SA analyses in this thesis are constrained by the assumption of static source profiles in PMF, which limits the model's ability to capture temporal evolution in

emissions and the atmospheric growth of particles. Consequently, NPF and subsequent particle growth, as well as diurnal or seasonal variations in source contributions, may be underestimated or misattributed in SA results.

- The findings of this thesis highlight the need for targeted regulatory and mitigation measures to reduce urban UFP exposure. Effective strategies should address not only road traffic emissions but also strengthen emission standards for residential heating, airports, and shipping, as well as tackle secondary formation processes driven by photochemical nucleation. Moreover, robust monitoring networks and standardised measurement approaches are essential to capture spatial and temporal variability and to evaluate policy effectiveness. This thesis has also contributed to the development of recommendations and service tools within the RI-URBANS project, aimed at guiding the harmonised measurement of UFPs and supporting the consistent application of SA methodologies across Europe. In addition, the results demonstrate the added value of implementing routine measurements of these emerging pollutants to enhance AQ assessment and public health protection.

7 FUTURE RESEARCH

This chapter provides recommendations for future research that could help address the knowledge gaps identified in this thesis:

- Although the inclusion of UFP measurements in the Ambient Air Quality Directive (EU) 2024/2881 ensures their implementation across Europe, further research is needed to consolidate the scientific basis linking UFP exposure to adverse health effects. The epidemiological evaluation of the association between UFP exposure and health effects will be essential to support future policy development. These studies should therefore use harmonised and quality-assured UFP datasets to assess both short- and long-term health outcomes with greater accuracy and comparability. Measurements below 10 nm typically exhibit higher uncertainties and lower inter-instrument comparability, which may undermine the robustness of epidemiological studies. Consequently, future health studies should rely on harmonised PNSD and PNC data with a lower size limit of 10 nm and no restriction on the upper size limit, as required by the Directive, ensuring consistency across monitoring networks and over time.
- While harmonised datasets with a lower size limit of 10 nm are most suitable for health-related applications, measurements extending below this threshold are essential for advancing the understanding of atmospheric and climate-related processes. In particular, particles smaller than 10 nm play a critical role in NPF, CCN activity, and other microphysical processes influencing climate. Future research should therefore promote the integration of Particle Size Magnifiers (PSMs) with SMPSs to achieve continuous and size-resolved measurements from 1 nm upwards. Such combined instrumentation would enable the detailed characterisation of nucleation and early particle growth dynamics, thereby improving our understanding of UFP formation and their subsequent atmospheric evolution. Furthermore, specific studies on the health effects of <10 nm UFPs should also be carried out, as information on this issue is currently lacking.

- Detailed knowledge of the chemical composition of freshly formed clusters is essential for improving the understanding the causes of NPF. The application of high-resolution mass spectrometric techniques, such as the Chemical Ionisation Atmospheric Pressure interface Time of Flight Mass Spectrometer (CI-APi-ToF), for particles below 10 nm can help identify precursor vapours and elucidate their role in particle formation and growth.
- Understanding which fraction of UFPs, solid or total, is most relevant for health outcomes remains an open question. Future research should include long-term measurements using instruments such as the Tandem Differential Mobility Analyser (TDMA) to measure both solid and total PNCs simultaneously. Comparing these fractions in epidemiological studies will help determine whether health effects are better correlated with solid particles or total particles, thereby providing critical information for targeted AQ monitoring.
- Improvements in SA techniques are needed to better capture the temporal evolution and growth of UFPs. While dispersion normalisation has proven valuable in separating emission-driven changes from meteorological variability, new methodologies that account for dynamic source profiles, particle growth, and temporal variations would enhance the accuracy of SA results.
- Systematic monitoring at multiple locations between airports and nearby cities can help quantify spatial gradients and distinguish primary emissions from secondary nucleation processes derived from SVOCs. Such studies will provide a detailed understanding of how airport emissions affect both large urban populations and smaller communities in close proximity.
- Characterising the transfer of UFPs from outdoor to indoor environments is also essential for accurately assessing human exposure. Research should investigate not only the infiltration of outdoor UFPs into indoor spaces but also the potential for NPF triggered by the infiltration of outdoor O₃ and the highly concentrated indoor VOCs, to improve exposure assessments and inform strategies to mitigate indoor UFP levels. Furthermore, the levels, sources, and abatement strategies for indoor UFPs should be investigated, as people spend approximately 85% of their time in indoor environments.

8 SCIENTIFIC CONTRIBUTIONS

8.1 Articles in peer-reviewed scientific journals

8.1.1 Publications included in this thesis

Trechera, P., **Garcia-Marlès, M.**, Liu, X., Reche, C., Perez, N., Savadkoohi, M., Beddows, D., Salma, I., Vörösmarty, M., Casans, A., Casquero-Vera, J.A., Hueglin, C., Marchand, N., Chazeau, B., Gille, G., Kalkavouras, P., Mihalopoulos, N., Ondracek, J., Zíková, N., Niemi, J.V., Manninen, H.E., Green, D.C., Tremper, A.H., Norman, M., Vratolis, S., Eleftheriadis, K., Gomez-Moreno, F.J., Alonso-Blanco, E., Gerwig, H., Wiedensohler, A., Weinhold, K., Merkel, M., Bastian, S., Petit, J.E., Favez, O., Crumeyrolle, S., Ferlay, N., Dos Santos, S.M., Putaud, J.P., Timonen, H., Lampilahti, J., Asbach, C., Wolf, C., Kaminski, H., Altug, H., Hoffmann, B., Rich, D. Q., Pandolfi, M., Harrison, R.M., Hopke, P.K., Petäjä, T., Alastuey, A., Querol, X., 2023. Phenomenology of ultrafine particle concentrations and size distribution across urban Europe. *Environment International*, 172, 107744. <https://doi.org/10.1016/j.envint.2023.107744>

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8.1.2 Co-author publications

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8.2 Conference presentations

Garcia-Marlès, M., Alastuey, A., Querol, X., Hopke, P.K., 2023. Source apportionment of ultrafine particle size distributions in urban Europe. European Geosciences Union (EGU) General Assembly, April 2023, Vienna, Austria

Trechera, P., **Garcia-Marlès, M.**, Alastuey, A., Querol, X., 2023. Phenomenology of ultrafine particle concentrations and size distribution across urban Europe. European Geosciences Union (EGU) General Assembly, April 2023, Vienna, Austria

Garcia-Marlès, M., Alastuey, A., Querol, X., Hopke, P.K., 2023. Source apportionment of ultrafine particle size distributions in urban Europe. European Aerosol Conference (EAC), September 2023, Málaga, Spain

Dos Santos, V.N., Rivas, I., **Garcia-Marlès, M.**, Pandolfi, M., Alastuey, A., Kalkavouras, P., Mihalopoulos, N., Grivas, G., Reche, C., Pérez, N., Salma, I., Vörösmarty, M., Bastian, S., Casans, A., Casquero-Vera, J.A., Petäjä, T., Lampilahti, J., Timonen, H., Weinhold, K., Wiedensohler, A., Green, D.C., Tremper, A.H., Gómez-Moreno, F.J., Alonso-Blanco, E., Hueglin, C., Petit, J.E., Favez, O., Altug, H., Hofmann, B., Savadkoohi, M., Liu, X., Ballester, J., Harrison, R.M., Hoek, G., Basagaña, X., Querol, X., 2024. Short-term effects of particles on mortality. European Geosciences Union (EGU) General Assembly, April 2024, Vienna, Austria

Dos Santos, V.N., Rivas, I., **Garcia-Marlès, M.**, Pandolfi, M., Alastuey, A., Kalkavouras, P., Mihalopoulos, N., Grivas, G., Reche, C., Pérez, N., Salma, I., Vörösmarty, M., Casans, A., Casquero-Vera, J.A., Petäjä, T., Lampilahti, J., Timonen, H., Gómez-Moreno, F.J., Alonso-Blanco, E., Hueglin, C., Ballester, J., Harrison, R.M., Hoek, G., Basagaña, X., Querol, X., 2024. Short-term effects of particle number size

distribution modes on health. International Society for Environmental Epidemiology (ISEE) Young, June 2024, Rennes, France

Garcia-Marlès, M., Lara, R., Alastuey, A., Petäjä, T., Querol, X., 2024. Ultrafine particles in urban Europe: results from RI-URBANS. Meeting on Aerosol Science and Technology (RICTA), June 2024, A Coruña, Spain

Querol, X., Alastuey, A., Pandolfi, M., Petäjä, T., **Garcia-Marlès, M.**, Liu, X., Lara, R., Savadkoohi, M., Salameh, T., Dufresne, M., Sauvage, S., Uzu, G., 2024. Advanced air quality parameters in the new air quality directive: the RI-URBANS/ACTRIS' view. Meeting on Aerosol Science and Technology (RICTA), June 2024, A Coruña, Spain

Garcia-Marlès, M., Lara, R., Alastuey, A., Petäjä, T., Querol, X., 2024. Inter-annual trends of ultrafine particles in urban Europe. European Aerosol Conference (EAC), August 2024, Tampere, Finland

Querol, X., Amato, F., Salameh, T., Uzu, G., Daellenbach, K., Via, M., Savadkoohi, M., **Garcia-Marlès, M.**, Petäjä, T., Timonen, H., Pandolfi, M., Alastuey, A., de la Rosa, J., Liu, X., Hopke, P.K., 2025. RI-URBANS: Source apportionment of different pollutants in urban Europe. European Geosciences Union (EGU) General Assembly, April 2025, Vienna, Austria

8.3 Project reports

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APPENDIX

Supplement of paper I

Phenomenology of ultrafine particle concentrations and size distribution across urban Europe

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Table S1. Data coverage of UFP-PNSD, range, range corrected, hours available, percentage of hours available and period. Furthermore, available data coverage of BC, PM, gaseous pollutants and meteorological parameters. BC, Black Carbon; PM, Particulate Matter; UB, Urban background; TR, Traffic; SUB, Suburban background; RG, Regional background; ND, Not-determined; T, Temperature; RH, Relative humidity; P, Pressure; RAD, Radiation; ws, Wind speed; WD, Wind direction.

Station	Range (nm)	Range corrected (nm)	Hours	% hours	Period	BC	Gases	PM	Meteorology
ATH_UB	10-467	No	16735	63.7	07/02/2017 – 30/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10	T, RH, P, RAD, ws, WD, RAIN
BCN_UB	11-478	12-478	20180	76.8	01/01/2017 – 31/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5, 1	T, RH, P, RAD, ws, WD, RAIN
BIR_UB	12-552	13-552	3818	14.5	08/07/2019 – 31/12/2019	Yes	NO, NO ₂ , O ₃	10, 2.5, 1	T, RH, P, RAD
BUD_UB	6-994	11-816	24360	92.7	28/01/2017 – 31/12/2019	ND	ND	ND	T, RH, P, RAD, ws, WD
DRE_UB	10-800	No	18994	72.3	01/01/2017 – 31/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃	10	T, RH, RAD, ws, WD
GRA_UB	11-496	No	21418	81.5	01/01/2017 – 31/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10	T, RH, P, RAD, ws, WD
HEL_UB	3-891	10-794	26254	99.9	01/01/2017 – 30/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5	T, RH, P, RAD, ws, WD
LAN_UB	10-544	No	21769	82.8	16/01/2017 – 31/12/2019	ND	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5, 1	T, RH, P, RAD, ws, WD
LEI_UB	10-800	No	23457	89.3	10/01/2017 – 30/12/2019	Yes	ND	ND	ND
LND_UB	17-604	No	13690	52.1	01/01/2017 – 31/12/2018	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5, 1	T, RH, P
LND2_UB	17-604	No	5485	20.9	28/11/2018 – 19/12/2019	Yes	NO, NO ₂ , O ₃	10, 2.5, 1	T, RH, P
MAD_UB	10-661	15-661	11265	42.9	01/01/2017 – 31/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5	T, RH, P, RAD, ws, WD, RAIN
MAR_UB	15-661	15-661	4566	17.4	13/11/2018 – 31/05/2019	Yes	SO ₂ , NO, NO ₂ , O ₃	10, 2.5, 1	T, ws, WD
MUL_UB	14-496	No	24170	92.0	01/01/2017 – 31/12/2019	Yes	NO, NO ₂	10, 1	T, RH, RAD, ws, WD, RAIN
ROC_UB	11-470	No	23748	90.4	06/01/2017 – 31/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	2.5	T, RH, P
ZUR_UB	11-478	17-478	9011	34.3	01/01/2017 – 13/11/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5	T, RH, P, RAD, ws, WD, RAIN
DRE_TR	5-800	10-800	20414	77.7	18/01/2017 – 31/12/2019	Yes	NO, NO ₂ , O ₃	10	T, RH, RAD, ws, WD
HEL_TR	6-800	11-800	18639	70.9	01/01/2017 – 05/06/2019	Yes	NO, NO ₂ , O ₃ , CO	10, 2.5	T, RH, P, ws, WD
LEI_TR	10-800	No	18165	69.1	22/11/2017 – 31/12/2019	Yes	SO ₂ , NO, NO ₂	10	T, RH, P, RAD, ws, WD
LEI2_TR	10-800	No	25731	97.9	01/01/2017 – 31/12/2019	Yes	ND	ND	ND
LND_TR	17-604	No	12209	46.5	01/01/2017 – 14/05/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5, 1	T, RH, ws, WD
STO_TR	10-410	No	12417	47.2	01/01/2017 – 17/09/2018	Yes	NO, NO ₂ , O ₃ , CO	10, 2.5, 1	T, RH, P, RAD, ws, WD, RAIN
BUD_TR	ND	ND	ND	ND	28/01/2017 – 31/12/2019	ND	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5	ND
ATH_SUB	10-550	No	19525	74.3	02/01/2017 – 31/12/2019	Yes	NO, NO ₂ , O ₃	10, 2.5	T, RH, P, RAD, ws, WD, RAIN
LIL_SUB	16-594	20-594	13306	50.6	04/07/2017 – 31/12/2019	Yes	NO, NO ₂ , O ₃	10, 2.5	T, RH, P, RAD, ws, WD, RAIN
PAR_SUB	11-792	11-792	24115	91.8	01/01/2017 – 31/12/2019	Yes	NO, NO ₂ , O ₃ , CO	10, 2.5, 1	T, RH, P, RAD, ws, WD, RAIN
PRA_SUB	9-519	10-519	22612	86.0	01/01/2017 – 31/12/2019	ND	SO ₂ , NO, NO ₂ , O ₃ , CO	10, 2.5	T, RH, P, ws, WD, RAIN
ISP_RB	10-800	No	25019	95.2	01/01/2017 – 31/12/2019	Yes	SO ₂ , NO, NO ₂ , O ₃ , CO	10	T, RH, P, RAD, ws, WD, RAIN

Table S2. Averaged levels of the atmospheric gaseous pollutants. UB, Urban background; TR, Traffic; SUB, Suburban background; RG, Regional background; ND, Not-determined.

Station	NO ₂ µg m ⁻³	NO µg m ⁻³	CO mg m ⁻³	SO ₂ µg m ⁻³	O ₃ µg m ⁻³	PM ₁₀ µg m ⁻³	PM _{2.5} µg m ⁻³	PM ₁ µg m ⁻³	BC µg m ⁻³
ATH_UB	30	18	0.37	7.0	74	36	19	ND	1.7
BCN_UB	29	7.6	0.30	1.7	50	20	13	8.6	1.5
BIR_UB	18	3.0	ND	ND	49	13	9.4	7.6	0.77
BUD_TR	46	33	0.66	5.0	27	37	16	ND	ND
DRE_UB	20	4.5	ND	3.2	46	18	ND	ND	0.74
GRA_UB	33	16	0.24	5.7	54	33	ND	ND	2.0
HEL_UB	6.5	2.8	0.23	0.68	56	10	4.2	ND	0.43
LAN_UB	23	6.2	0.26	0.95	46	13	11	9.1	ND
LEI_UB	ND	ND	ND	ND	ND	ND	ND	ND	0.88
LND_UB	31	11	0.18	2.0	42	14	8.8	8.0	0.85
LND2_UB	27	10	ND	ND	46	17	12	10	1.1
MAD_UB	19	5.8	0.21	3.9	67	16	9.2	ND	1.2
MAR_UB	29	8.2	ND	2.2	51	18	11	8.9	1.6
MUL_UB	23	7.1	ND	ND	ND	19	ND	12	0.91
ROC_UB	15	3.2	0.28	1.5	54	ND	6.2	ND	0.33
ZUR_UB	25	5.3	0.27	0.89	56	15	11	ND	0.48
DRE_TR	29	17	ND	ND	43	21	ND	ND	1.0
HEL_TR	32	24	0.22	ND	40	18	6.9	ND	1.0
LEI_TR	36	38	ND	1.8	ND	21	ND	ND	1.8
LEI2_TR	ND	ND	ND	ND	ND	ND	ND	ND	1.5
LND_TR	80	107	0.33	5.6	19	22	13	11	2.7
STO_TR	36	28	0.30	ND	58	24	9.0	6.3	0.99
ATH_SUB	13	1.7	ND	ND	82	18	11	ND	0.82
LIL_SUB	26	6.2	ND	ND	45	19	15	ND	1.0
PAR_SUB	11	2.5	0.16	ND	58	14	9.2	7.5	0.64
PRA_SUB	17	4.4	0.34	2.6	54	20	16	ND	ND
ISP_RB	5.4	5.2	0.08	0.35	51	25	ND	ND	1.5

Table S3. Patterns of UFP and behaviour with BC and wind direction in each site. UB, Urban background; TR, Traffic; SUB, Suburban background; RG, Regional background; ND, Not-determined; Nucl, Nucleation; Aitk, Aitken; Accum, Accumulation; ws, Wind speed; N, North; S, South; W, West; E, East; NW, North-west; NE, North-east; SW, South-west; SE, South-east.

Station	PNC peak at rush hours	Midday M_{10-25} peak and time	PNC seasonality	PNC & BC seasonality	Wind direction with UFP modes
ATH_UB	High at 6am, but higher in the evening	Light peak at 10am, minimum in summer	Lowest PNC in summer, highest in winter; Low at weekends	Parallel seasonally and daily	Nucl from NW with high ws; Aitk and Accum stagnation
BCN_UB	High in the morning and evening	Peak at 12pm; Higher in summer followed by spring	Similar patterns at each season; Lowest at weekends	High, except for the midday peak	Nucl from S and W; Aitk and Accum stagnation
BIR_UB	Low at 7am, high at 8pm	Light peak (in summer)	Lowest in winter and at weekends	Very similar	ND
BUD_UB	High peaks in the morning and evening	Very high at 11am (highest in spring); Higher Nucl than Aitk	Highest in autumn and spring; Lowest at weekends	ND	Nucl from SW and W with high ws; Aitk from S, also stagnation
DRE_UB	Yes, much higher in the evening	Yes, but in general low Nucl values	Highest PNC in summer; lowest at the weekends	High, except for the midday peak	Nucl and Aitk from NE; Accum a bit from E
GRA_UB	High peaks in the morning and evening	Light peak at 12pm (higher in summer)	Highest PNC in summer; lowest in winter and at weekends	Parallel seasonally and daily	Stagnation; Nucl from NW; Aitk
HEL_UB	Two peaks, but higher in the morning	Light at 11am in spring and summer	Lowest PNC in winter and at weekends	Hourly similar, but BC has a minimum peak in summer	Nucl from NW; Aitk and Accum stagnation
LAN_UB	Yes (morning), but high during day hours	Light at 3pm, but high during day hours	High values in summer; Nucl is the highest	ND	Nucl from NW (airport); Aitk stagnation; Accum from E
LEL_UB	Yes, higher in the evening	Yes (11am). Very high in summer	Lowest in winter and at weekends	BC low in summer, daily similar (but no midday peak)	ND
LND_UB	Yes, higher in the evening	Light peak, a bit high in summer	Highest in summer and autumn; lightly lower at the weekends	BC low in summer; but daily very similar	ND
LND2_UB	Yes, higher in the evening	Light peak, a bit high in summer	Highest in summer and autumn; lightly lower at the weekends	BC low in summer; but daily very similar	ND
MAD_UB	High peak in the morning and low in the evening	No peak (a bit in summer and winter), but rush hour peaks	Highest values in autumn and winter; Higher PNC in day hours	Parallel seasonally and daily	Air stagnation but Nucl mostly from SW
MAR_UB	High peaks	Only in spring (no summer data)	Highest in winter and autumn	Parallel seasonally and daily	Nucl from W; others from NE
MUL_UB	High in the morning and evening	Light peak at 12am, higher peak in summer	Highest PNC in spring and slightly lower in summer	BC low in summer; but daily very similar	Nucl from NW; Aitk from NW
ROC_UB	Yes, in the morning and late evening	Only in summer at 11am; Nucl has rush hour peaks.	Highest PNC in summer and autumn	Similar, but better without Nucl	ND
ZUR_UB	Yes, higher in the evening	No peak, very low Nucl values	Highest in autumn and spring; Nucl higher in winter	Parallel seasonally and daily	Nucl from SE-S with high ws; Aitk stagnation; Accum from E

Table S3. (Continued) Patterns of UFP and behaviour with BC and wind direction in each site. UB, Urban background; TR, Traffic; SUB, Suburban background; RG, Regional background; ND, Not-determined; Nucl, Nucleation; Aitk, Aitken; Accum, Accumulation; ws, Wind speed; N, North; S, South; W, West; E, East; NW, North-west; NE, North-east; SW, South-west; SE, South-east.

Station	PNC peak at rush hours	Midday N_{10-25} peak and time	PNC seasonality	PNC & BC seasonality	Wind direction with UFP modes
DRE_TR	Peak at 6am; much higher at 7pm	Very high (not in winter); around 11am	Lowest PNC in July and at weekends	High; except for midday peak	Nucl and Aitk from NE; Accum stagnation
HEL_TR	Peak only in the morning	Light at 1pm; and same patterns the three modes	No seasonality; but Nucl higher in winter	Very similar	From NW and stagnation; the three modes
LEI_TR	Low peaks in the morning and evening	Yes (11am); Very high in summer	Seasonally flat; Lowest at the weekends	BC low in summer; daily similar (but no midday peak)	Air stagnation and from E
LEI2_TR	Yes; low in the morning	Yes (11am); Very high in summer	High in summer; low at weekends	High; except for the midday peak	ND
LND_TR	High peaks	No peak; Very low Nucl values	Lowest in spring and at weekends	Few values of BC; difficult to see	From SW, S, SE, and stagnation
STO_TR	Light peaks	No peak; but higher values during day hours	Highest PNC in late summer; lowest in winter and weekends	Parallel seasonally and daily	N Nucl with high ws; SE-S Aitk-Accum and stagnation
ATH_SUB	Low in the morning; but very high in the evening	Yes (11am); Much Higher in spring, (winter and autumn)	Lowest PNC in July-August and weekends; highest in spring	Low; two high BC peaks at rush hours; morning and evening	NW Nucl with high ws; W Aitk-Accum and stagnation
LIL_SUB	High peaks	Light peak but high in summer	Highest in summer and lightly lower at weekends	Few values of BC; difficult to see	ND
PAR_SUB	High peak in the evening; low in the morning	Very light; Low values of Nucl	Highest PNC in summer; Lowest at weekends	BC low in summer; but daily very similar	From NE the three modes, Aitken stagnation.
PRA_SUB	Yes; higher in the evening	Yes (10am); Very high in summer	Highest in summer and very low in winter; Also low at weekends	ND	Nucl from W; Aitk and Accum from N, NE, E
ISP_RB	Low in the morning; High in the evening	Yes (11am); High in summer and spring	Highest PNC in winter; lower in summer but with higher Nucl	Similar; without Nucl; High values at night for both	Air stagnation and a bit from W

Table S4. PNC (N_{10-800}) (top) and N_{25-800} (bottom) correlations with other parameters. TR, Traffic site, UB, Urban background site, SUB, Suburban background site; RB, Regional background site.

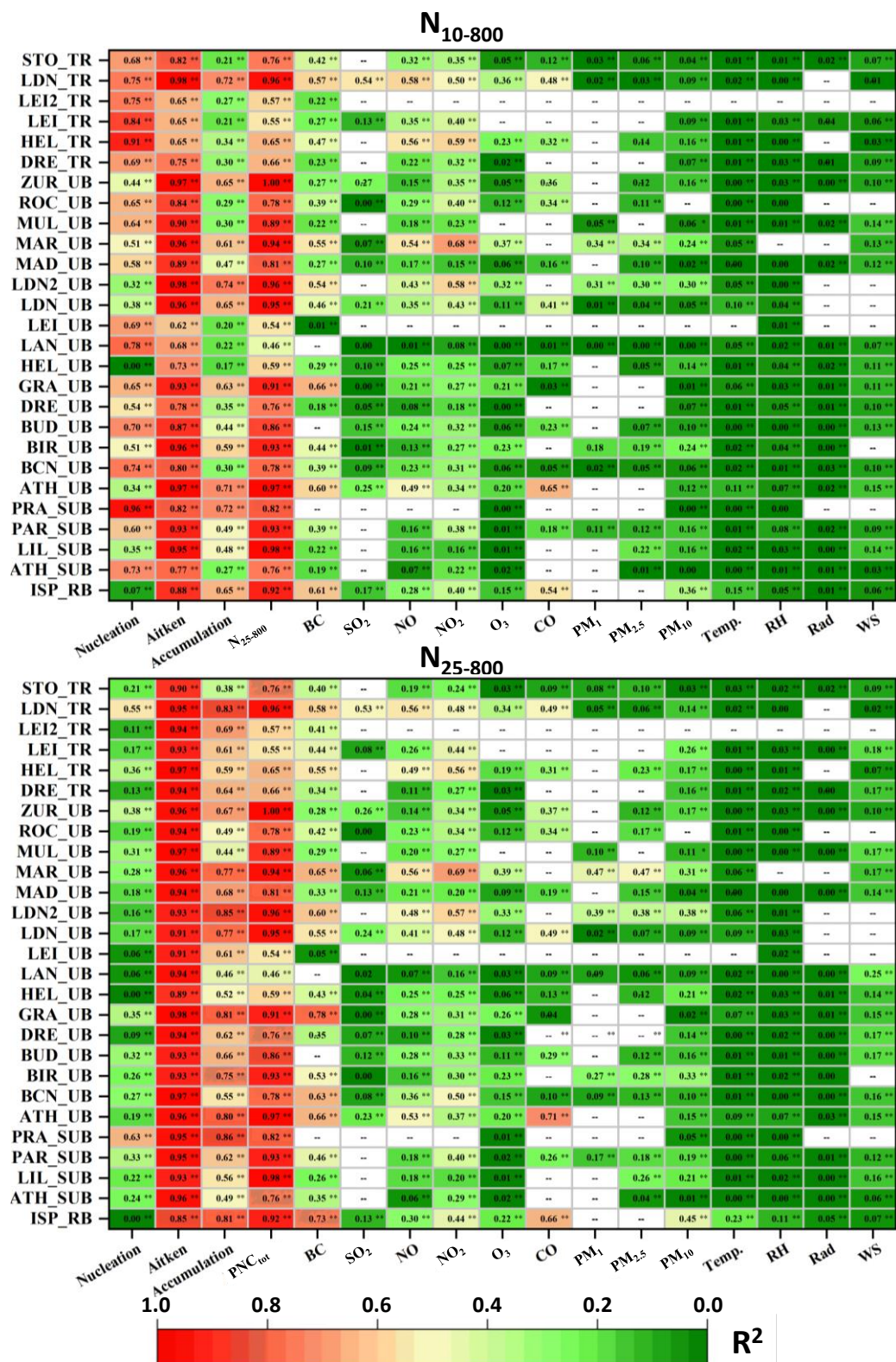


Table S5. Correlations of the Nucleation (N_{10-25}), Aitken (N_{25-100}) and Accumulation ($N_{100-800}$) modes concentrations with other parameters. TR, Traffic site, UB, Urban background site, SUB, Suburban background site; RB, Regional background site.

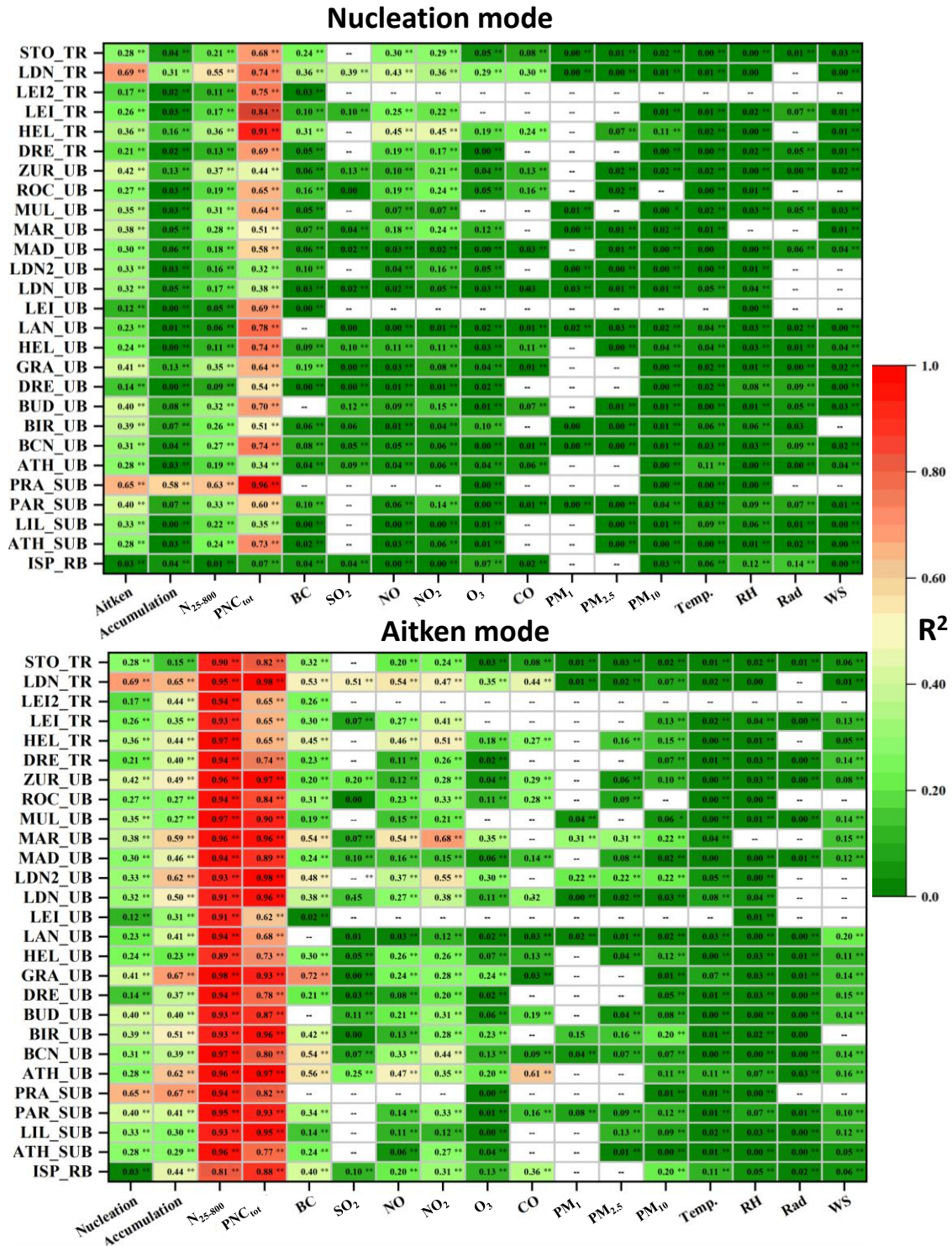


Table S5. (Continued)

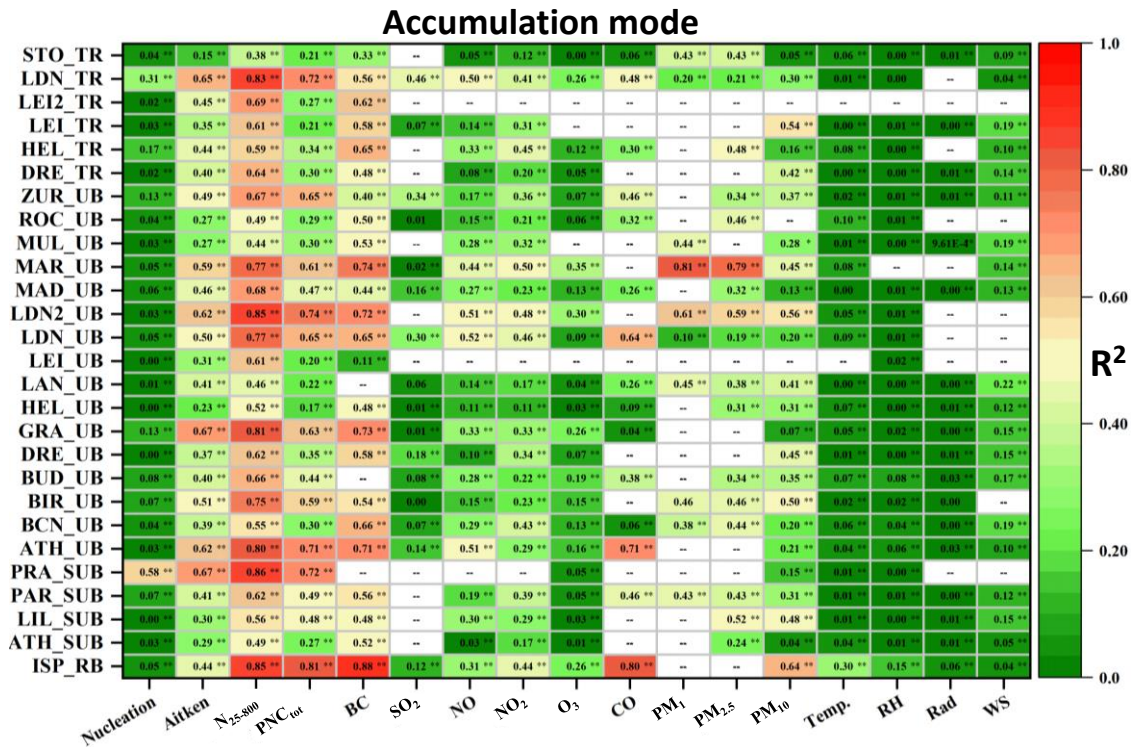


Table S6. PNC (N₁₀₋₈₀₀) and N₂₅₋₈₀₀ average correlations with other parameters. A positive correlation is indicated with the black number; a negative correlation is indicated with the red number. TR, Traffic site, UB, Urban background site, SUB, Suburban background site; RB, Regional background site.

R ²	PNC			N ₂₅₋₈₀₀		
	UB	TR	SUB+RB	UB	TR	SUB+RB
SO ₂	0.21	-	-	0.42	-	-
CO	0.21	0.04	0.00	0.21	0.36	0.08
NO	0.27	0.06	0.23	0.35	0.70	0.63
NO ₂	0.35	0.03	0.00	0.48	0.63	0.00
PM ₁₀	0.37	0.32	0.68	0.53	0.00	0.91
PM _{2.5}	0.40	0.05	0.77	0.49	0.77	0.91
PM ₁	0.10	-	-	0.12	-	-
O ₃	0.01	0.72	0.04	0.00	0.91	0.24
T	0.37	0.13	0.17	0.50	0.44	0.07
RH	0.22	0.01	0.00	0.39	0.30	0.07
RAD	0.18	0.76	0.16	0.31	0.82	0.14
WS	0.05	0.15	0.00	0.03	0.13	0.27

Figure S1. Meteorological parameters of the twenty-seven cities selected. T, Temperature; RAD, Solar Radiation; RH, Relative Humidity; ws, Wind Speed.

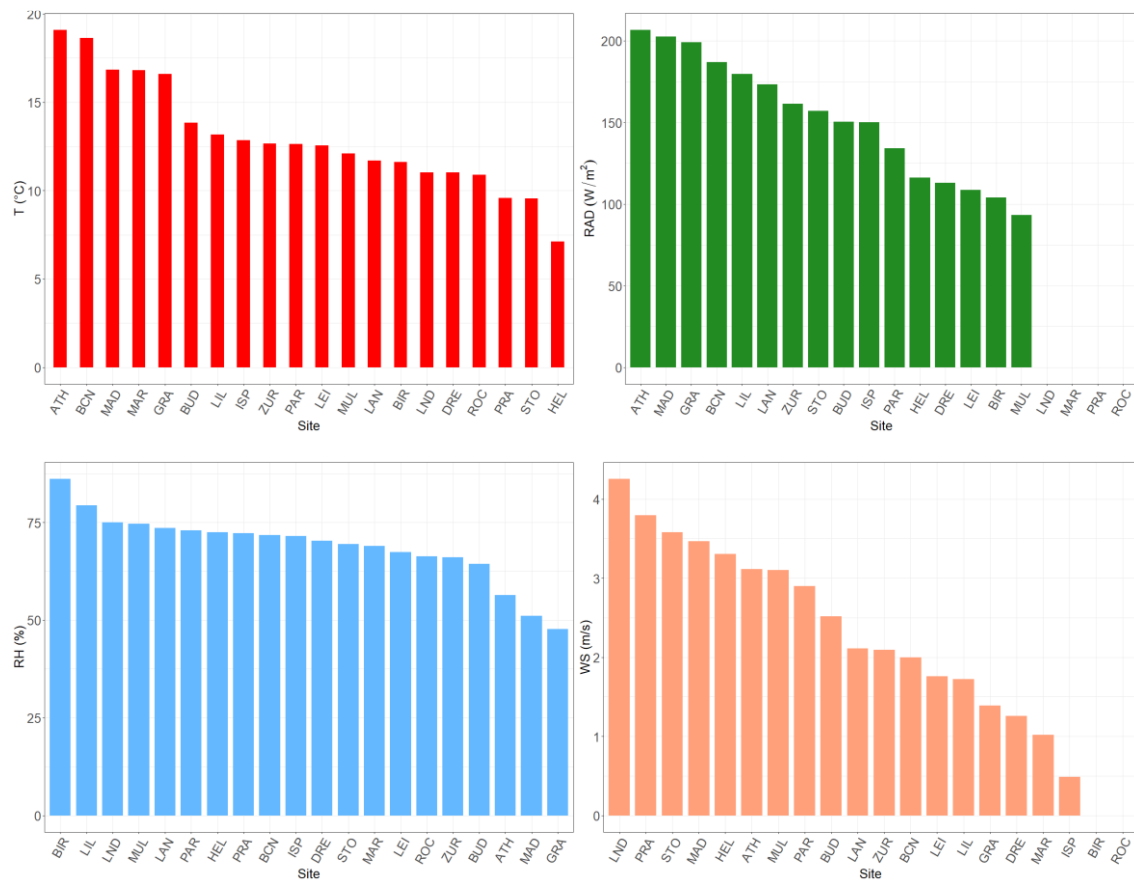


Figure S2. Representations of the twenty-seven UFP-PNSD datasets obtained. Top-left) All UFP-PNSD data as received. Top-right) Applied the ACTRIS and CEN recommendations (10-800 nm). Bottom-left) Incoherence measurements in the finer range for five datasets. Bottom-right) All datasets corrected following the ACTRIS and CEN recommendations used in this study.

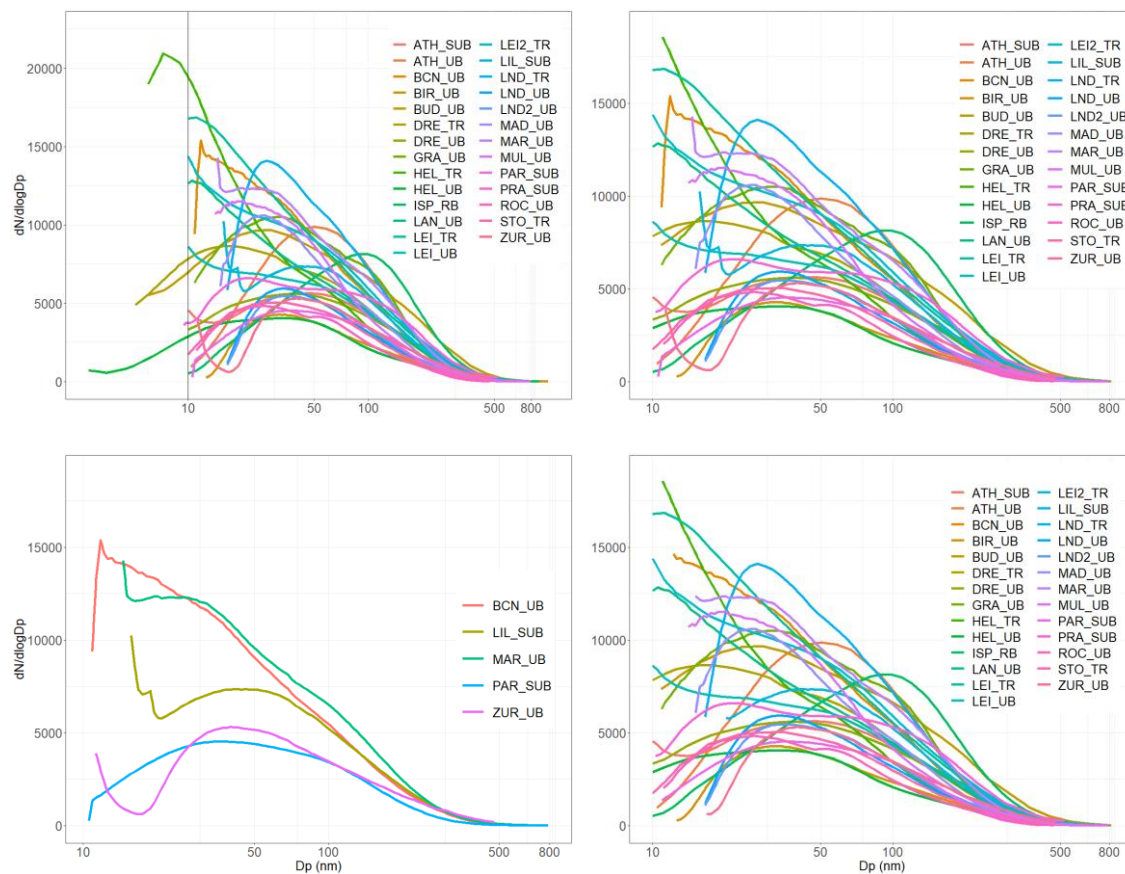


Figure S3. Correlation of 2017-2019 averaged concentrations of BC vs N_{10-25} , N_{25-100} and $N_{100-800}$ for all sites. The red sites in the graph of Nucleation are those with lower size detection limits of 15-20 nm.

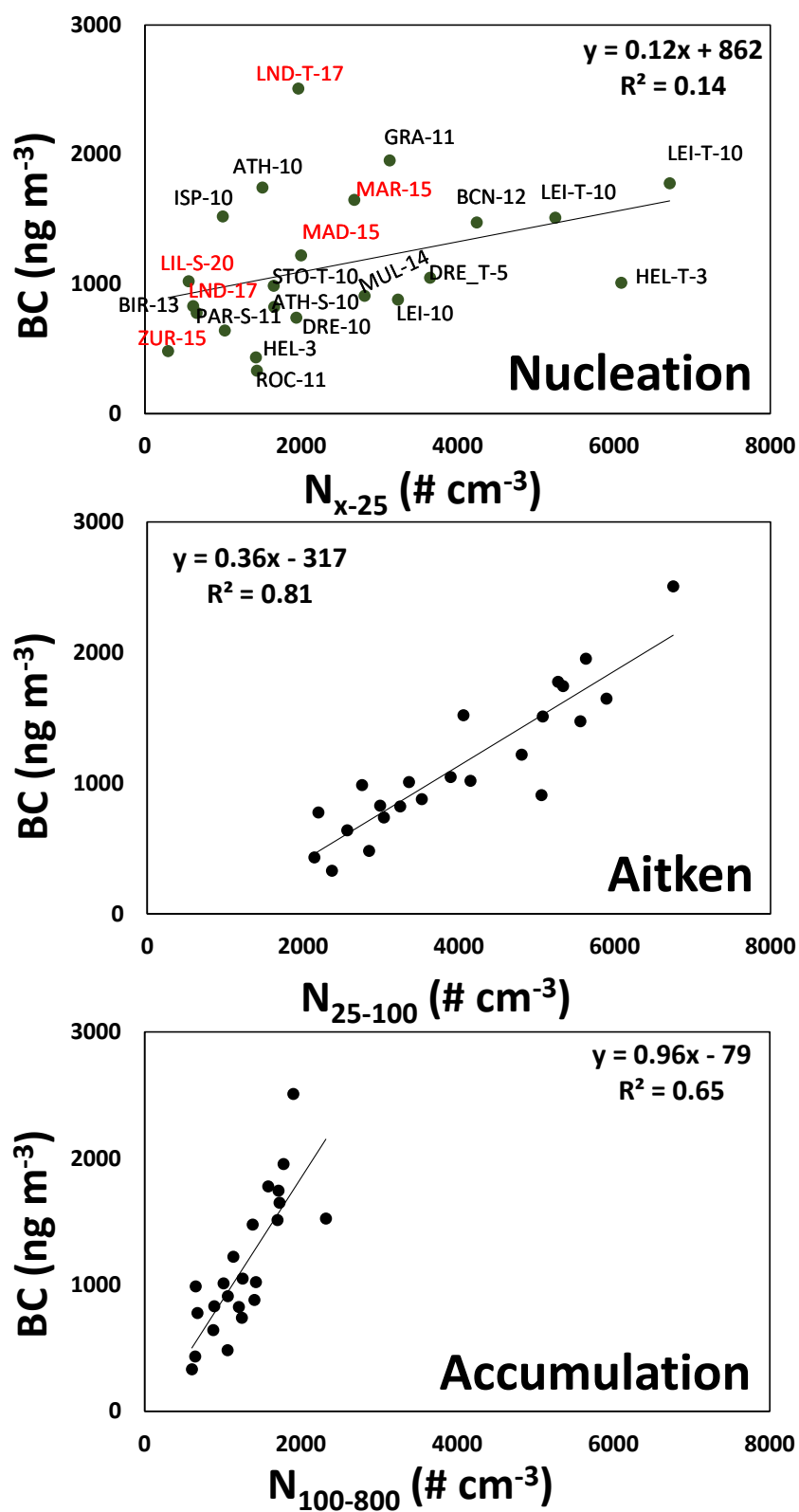
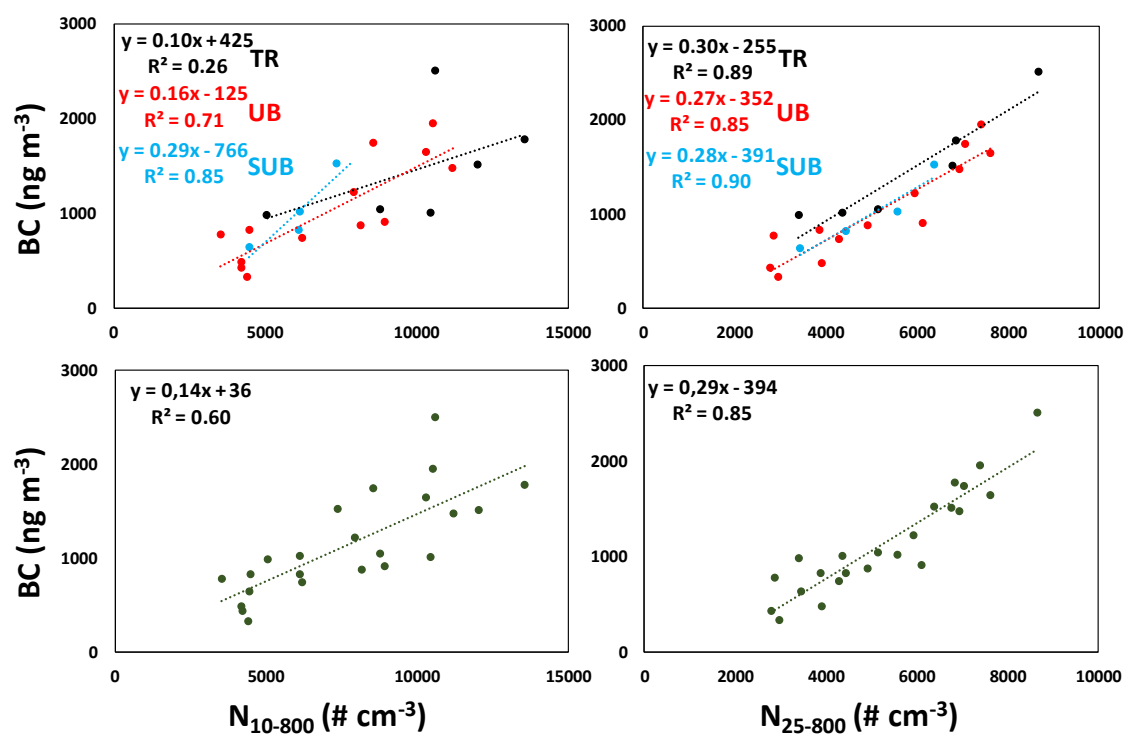


Figure S4. Correlation of 2017-2019 averaged concentrations of BC vs N_{10-800} and N_{25-800} for different type of sites (top) and for all sites (bottom).



Supplement of paper II

Inter-annual trends of ultrafine particles in urban Europe

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Table S1. Number of hours available of UFP-PNSD each year of the studied period for each site.

Site	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
ATH_UB	-	-	-	-	-	-	3038	5272	6968	4180	5587
BCN_UB	-	-	-	-	7460	6346	7316	7181	5331	8258	5345
BUD_UB	-	-	-	-	1177	8744	8387	4741	8099	7886	8377
DRE_UB	-	732	7980	7271	7757	6780	7824	7142	6148	6236	6610
HEL_UB	8698	8525	8210	8697	8484	8504	8722	8736	8562	8514	8623
LEC_UB	-	-	-	-	-	-	7768	5048	6359	7551	7559
LEI_UB	8638	8403	-	8268	8149	8444	8523	8266	7451	7451	8555
LND_UB	-	-	6477	6526	7934	7550	7311	7410	7183	5765	-
MAD_UB	3116	4928	6205	3820	6122	4232	5942	4997	3225	3421	4619
MUL_UB	6653	7952	8182	7849	8359	8760	7578	7425	7233	8219	8719
ROC_UB	7968	7608	8663	6335	8544	7520	8122	8496	8487	7584	7641
ZUR_UB	-	-	-	-	-	-	8182	7454	4829	3733	-
DRE_TR	4907	6108	7813	4019	7506	7710	8276	6754	5070	7287	7740
HEL_TR	-	-	-	-	-	-	7396	7532	8760	8760	3731
LEI_TR	-	3468	7878	7802	8119	8151	6069	7500	959	8614	8592
LND_TR	-	6615	6327	3548	7626	7227	7837	8119	5142	3879	3188
STO_TR	-	1705	7762	8392	8087	6249	5749	4916	6459	5957	-
ATH_SUB	-	5155	6873	7816	7711	8551	7520	7312	5955	5500	8070
PAR_SUB	-	-	-	2086	1300	4967	4436	6023	7036	8678	8368
PRA_SUB	-	-	-	5777	6179	7673	8041	3359	8759	8738	8760
IPR_RB	3903	8141	7933	7849	-	2958	-	5903	8415	8489	8115

ATH_UB

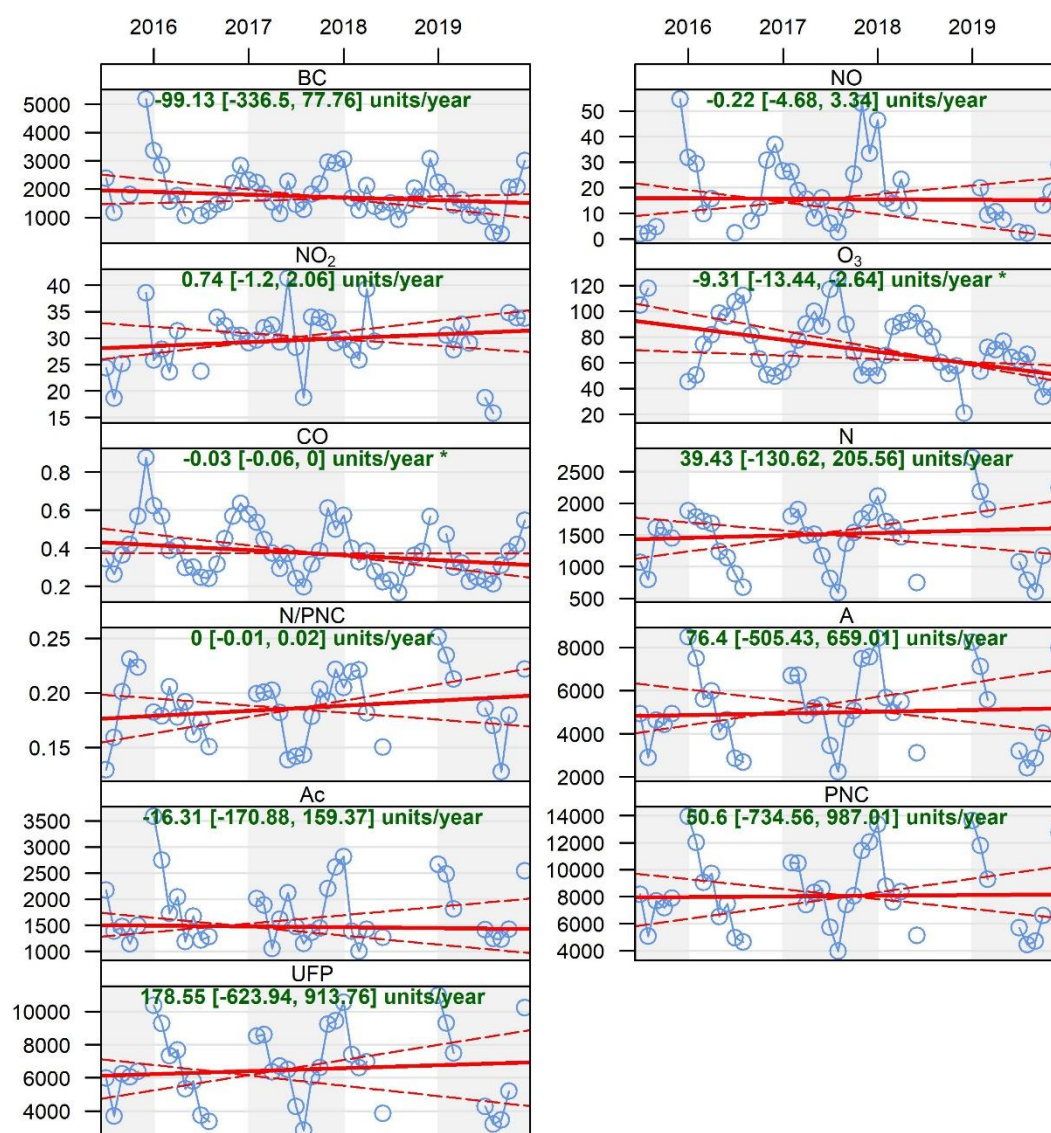


Figure S1. Trend analysis of all pollutants available in Athens urban background site (ATH_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

ATH_SUB

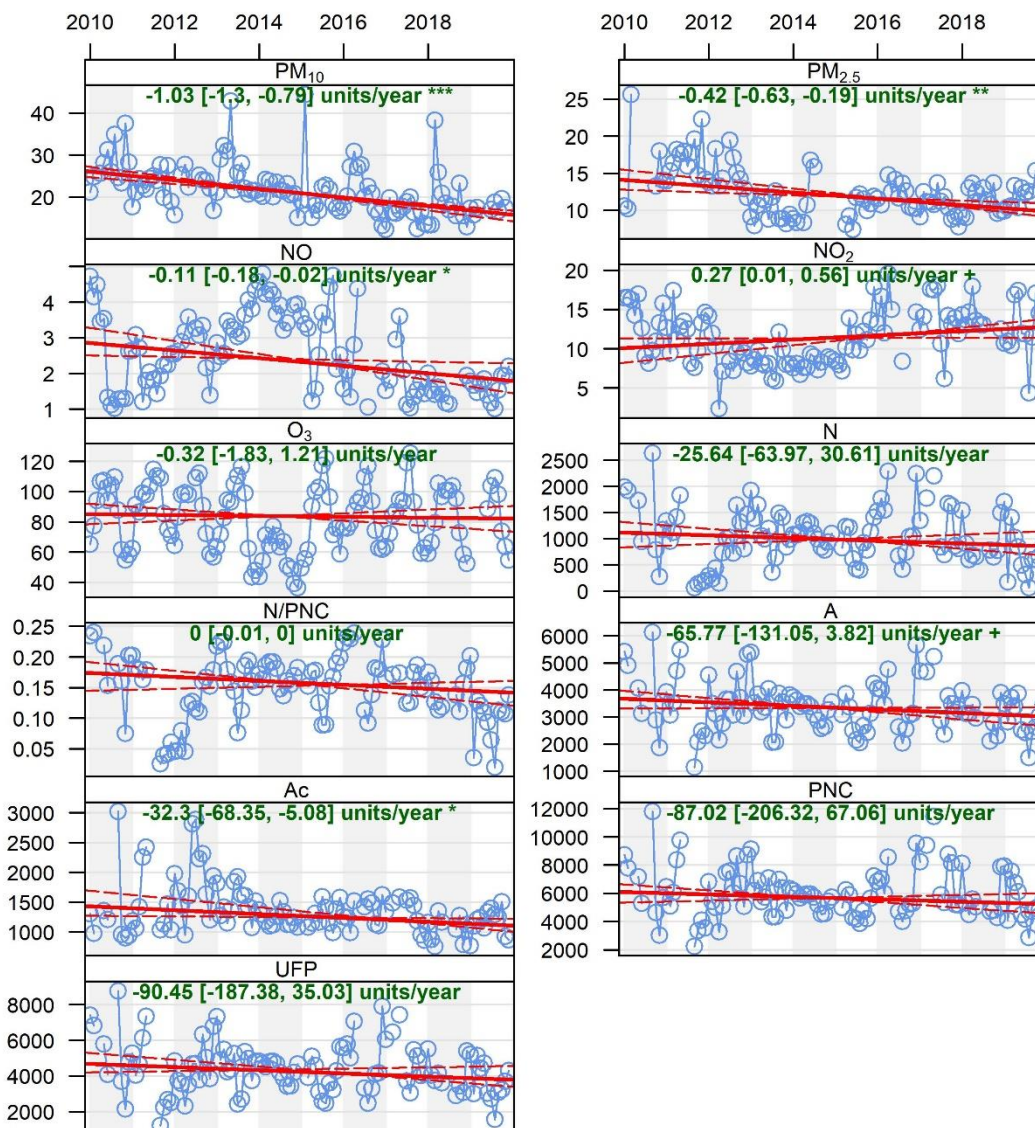


Figure S2. Trend analysis of all pollutants available in Athens suburban background site (ATH_SUB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

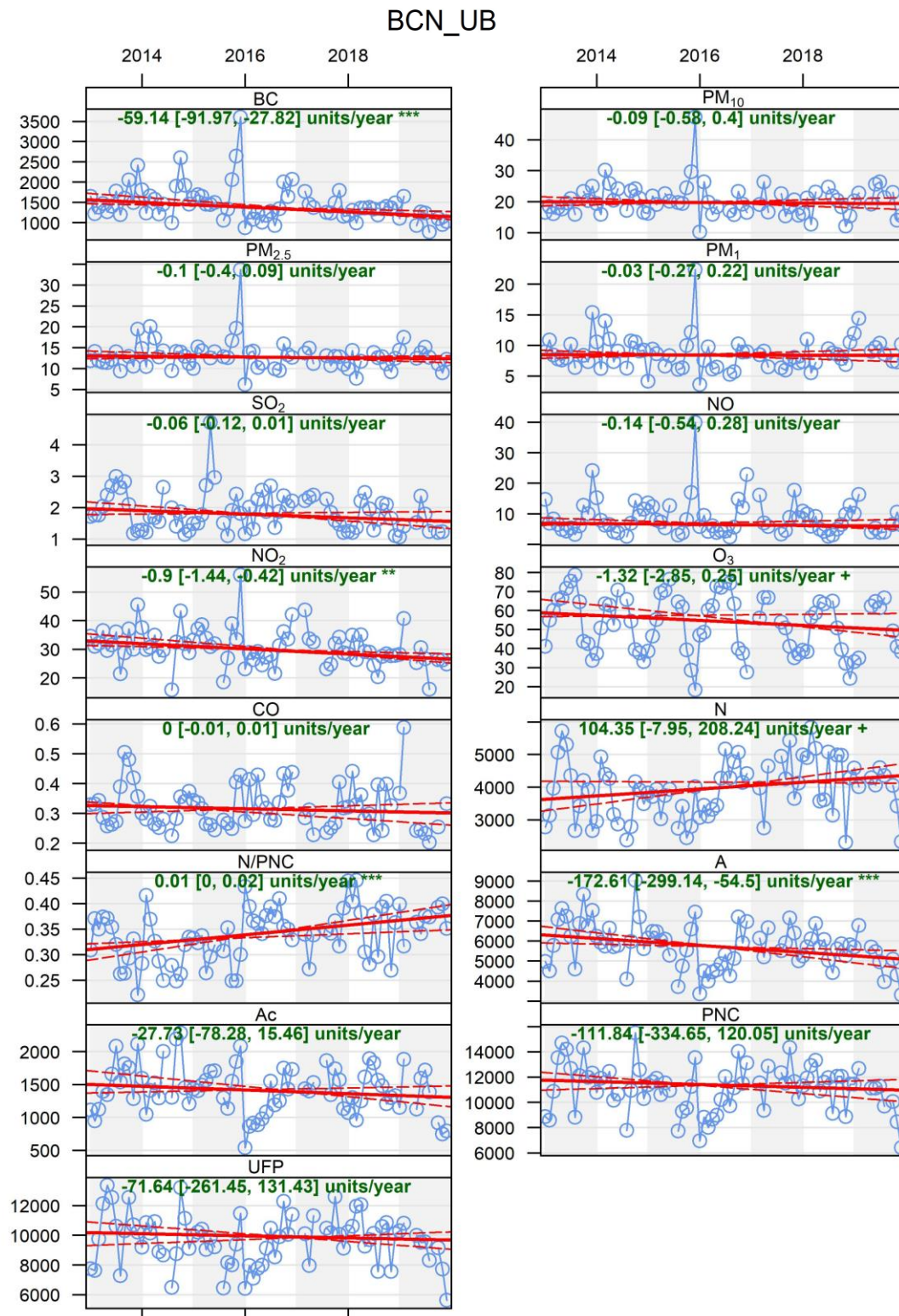


Figure S3. Trend analysis of all pollutants available in Barcelona urban background site (BCN_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

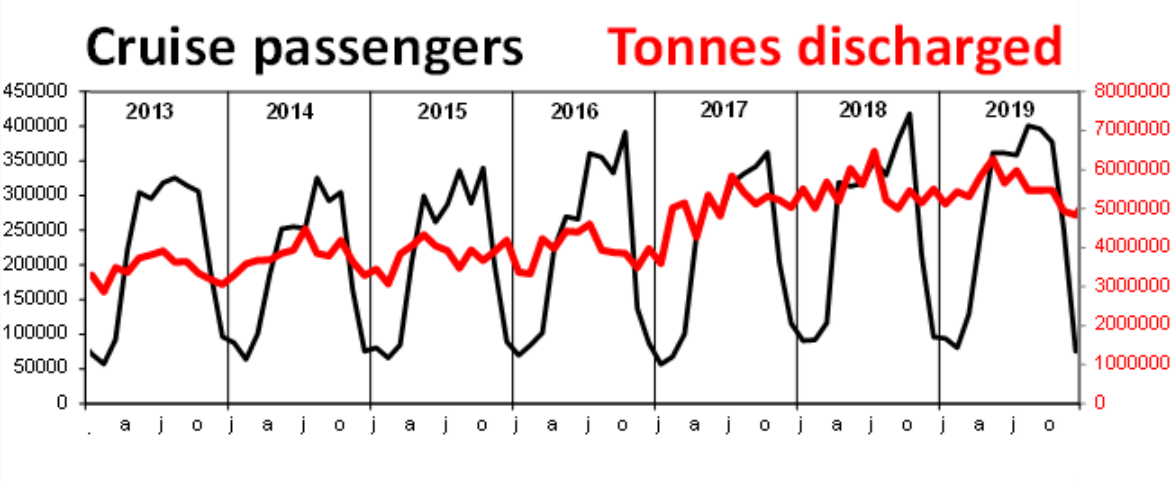


Figure S4. Monthly data on cruise passengers and tonnes of discharge in the harbour of Barcelona, according Port de Barcelona (2023).

BUD_UB

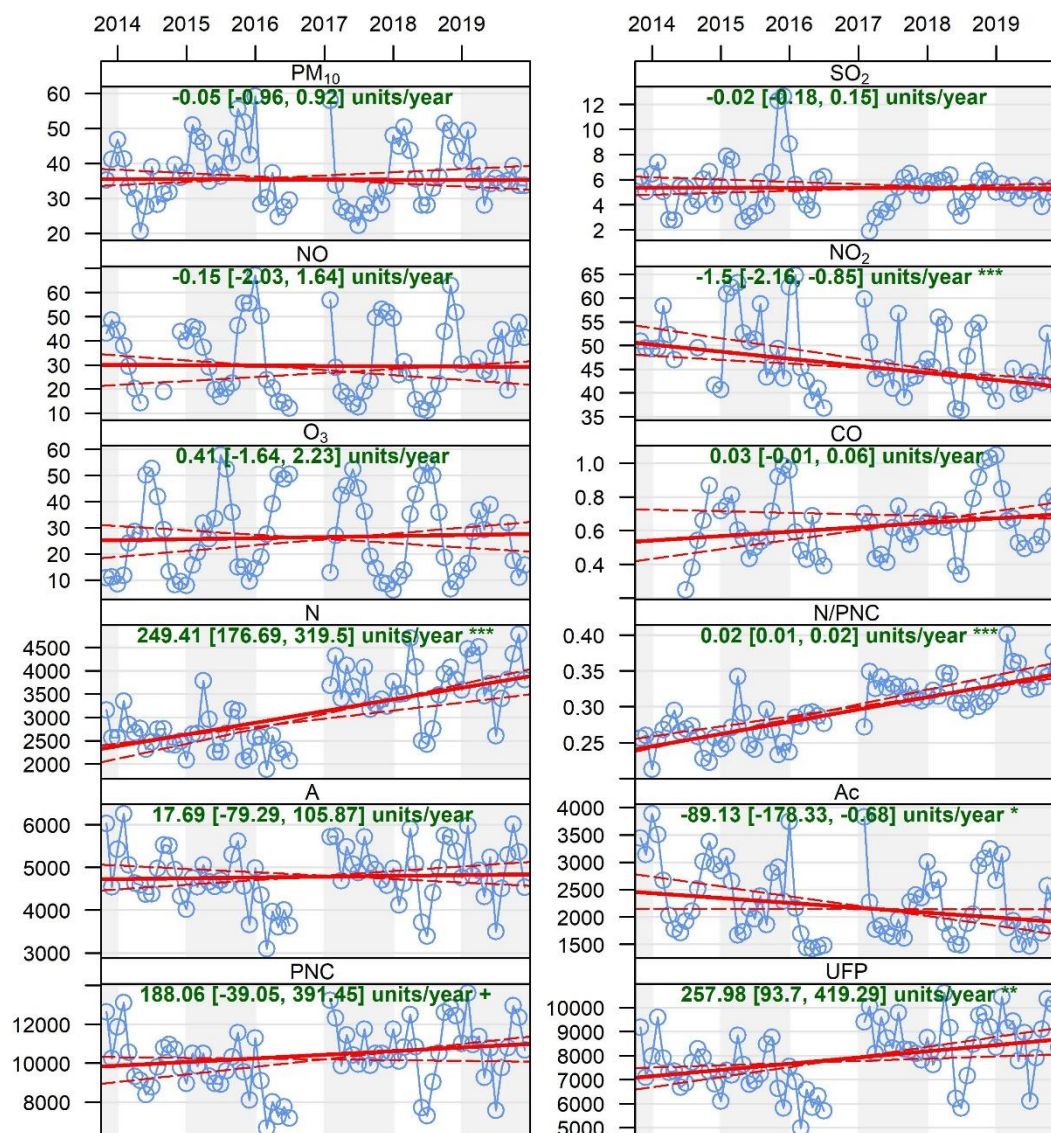


Figure S5. Trend analysis of all pollutants available in Budapest urban background site (BUD_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

DRE_UB

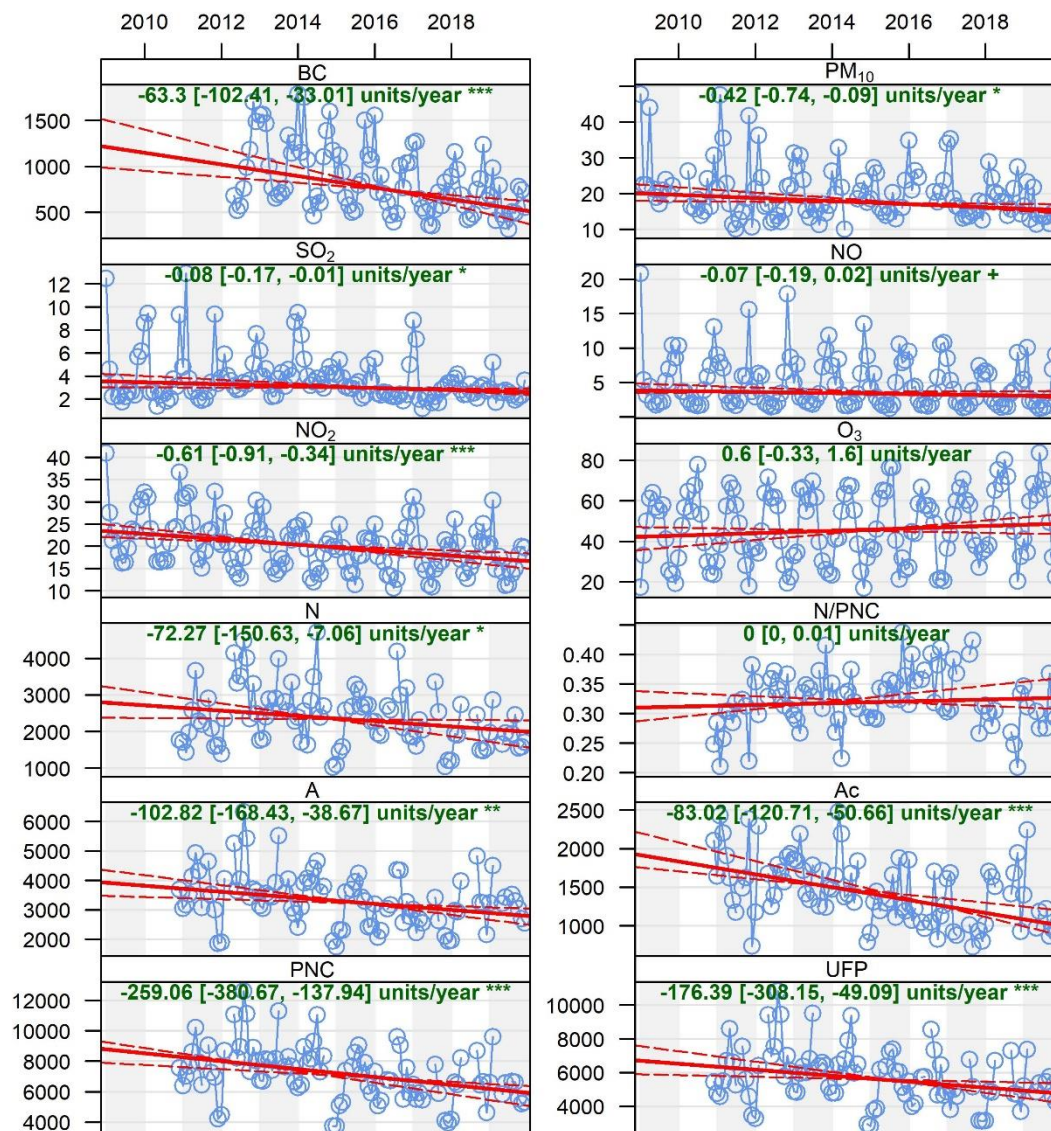


Figure S6. Trend analysis of all pollutants available in Dresden urban background site (DRE_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

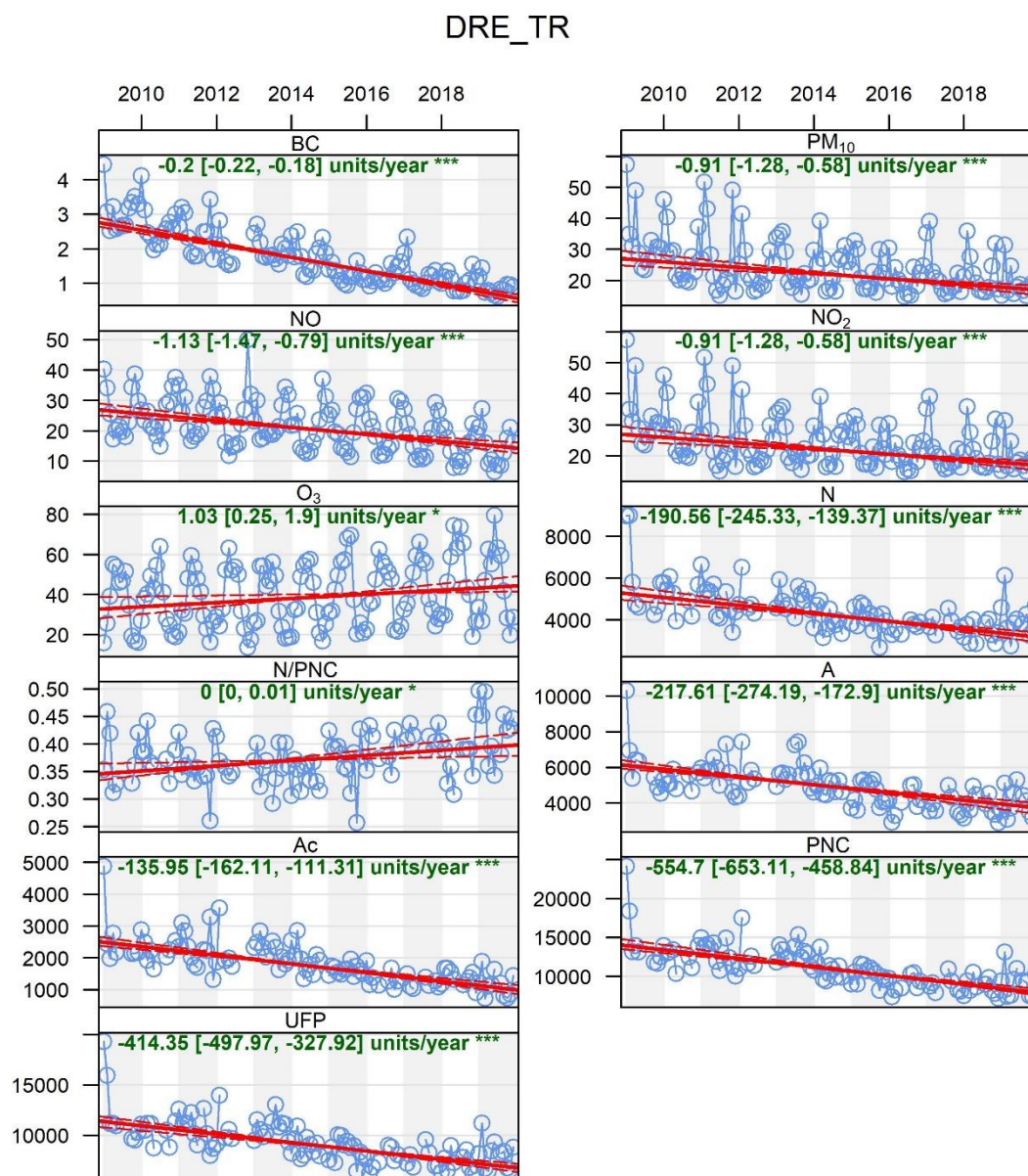


Figure S7. Trend analysis of all pollutants available in Dresden traffic site (DRE_TR). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

HEL_UB

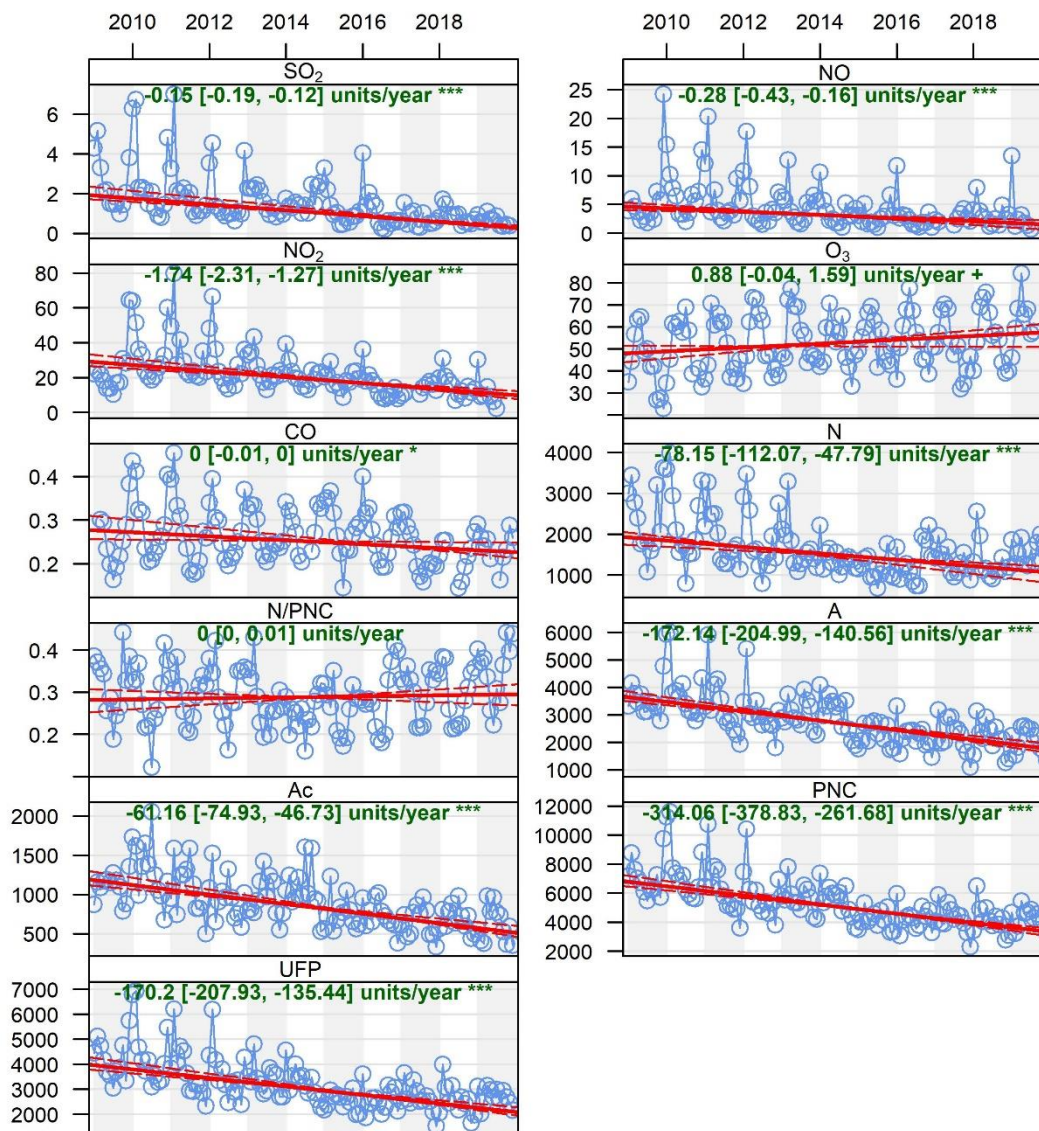


Figure S8. Trend analysis of all pollutants available in Helsinki urban background site (HEL_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

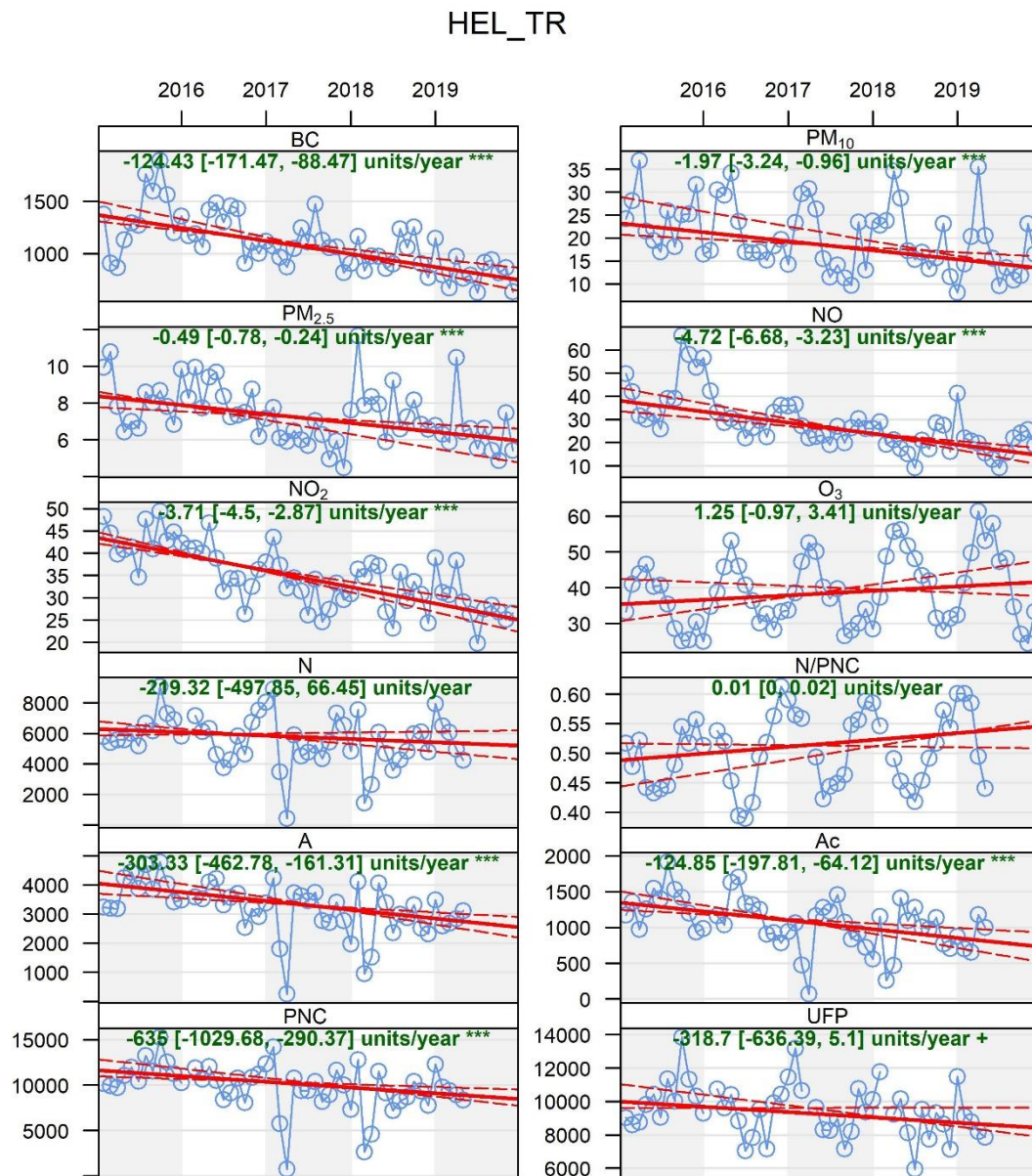


Figure S9. Trend analysis of all pollutants available in Helsinki traffic site (HEL_TR). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

IPR_RB

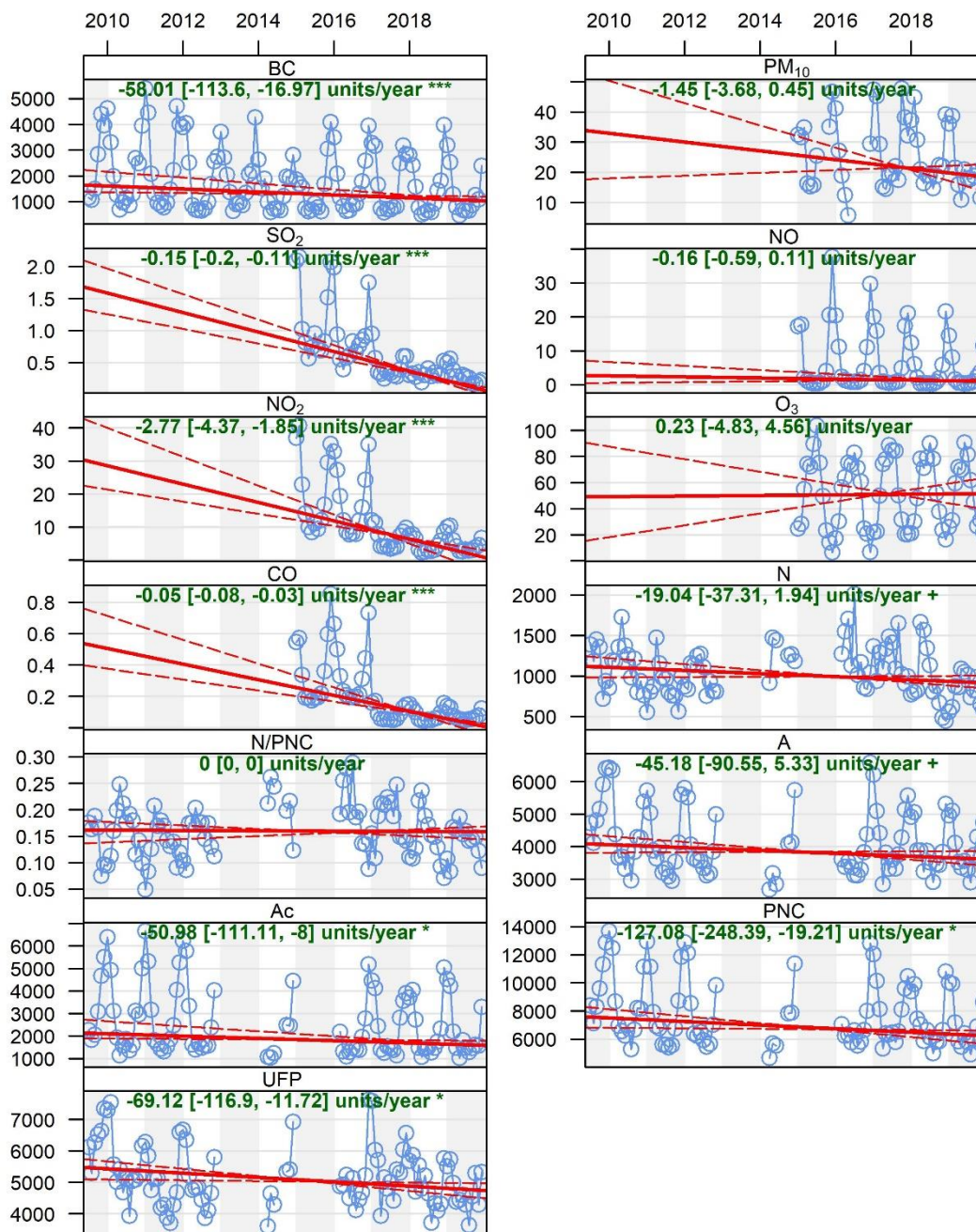


Figure S10. Trend analysis of all pollutants available in Ispra regional background site (ISP_RB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

LEC_UB

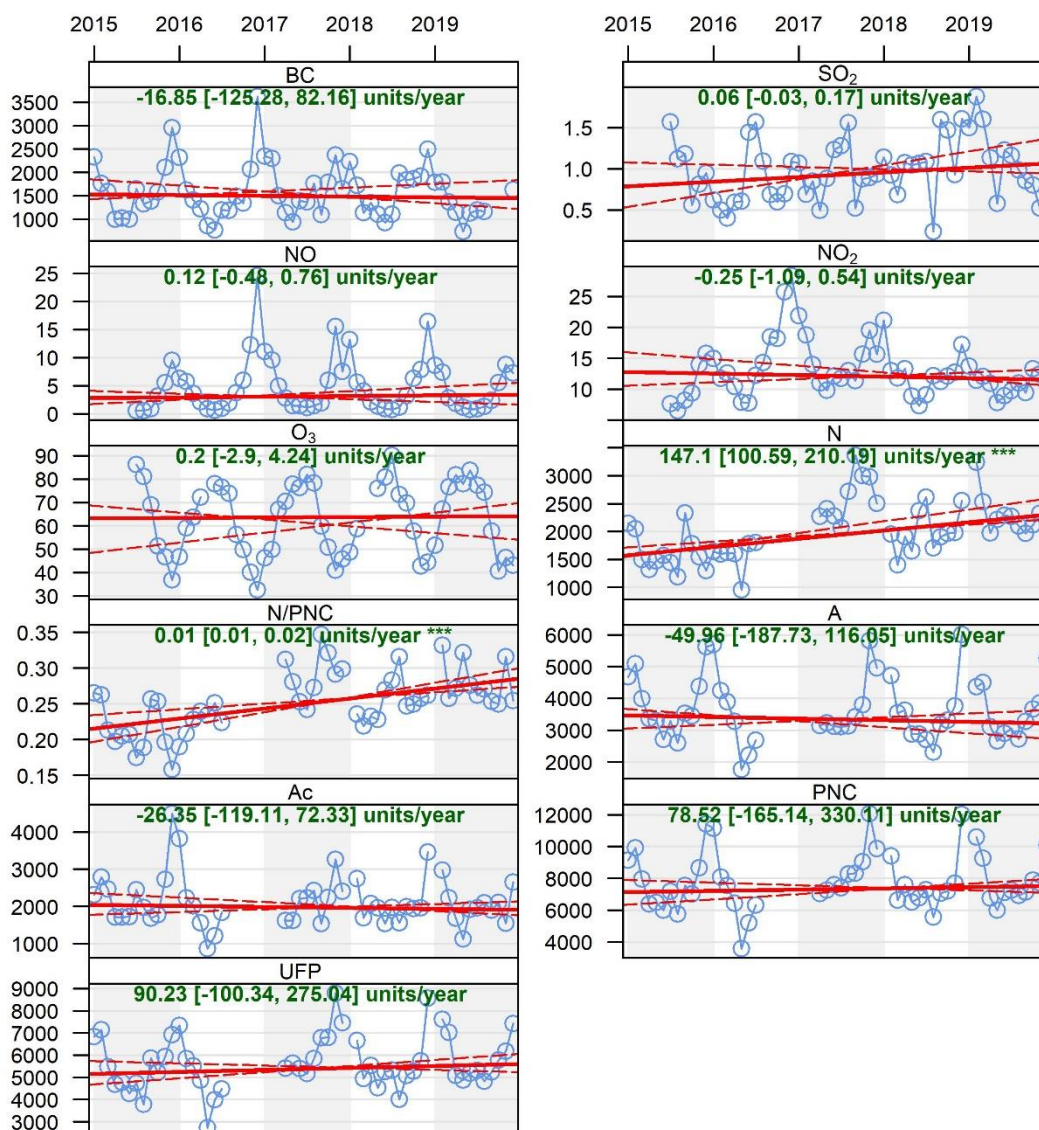


Figure S11. Trend analysis of all pollutants available in Lecce urban background site (LEC_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

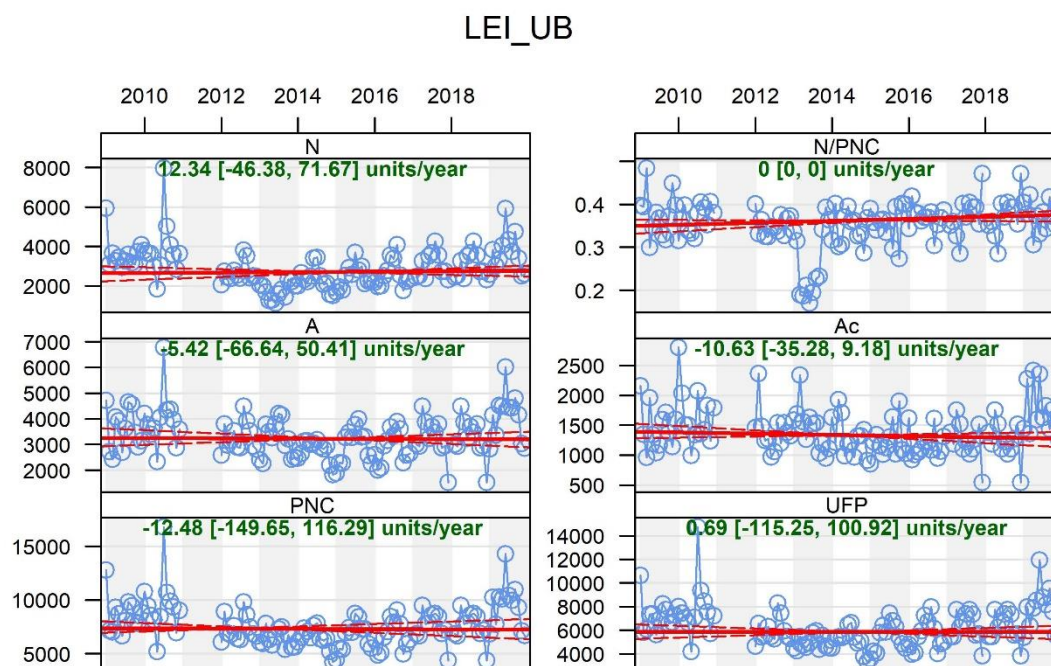


Figure S12. Trend analysis of all pollutants available in Leipzig urban background site (LEI_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

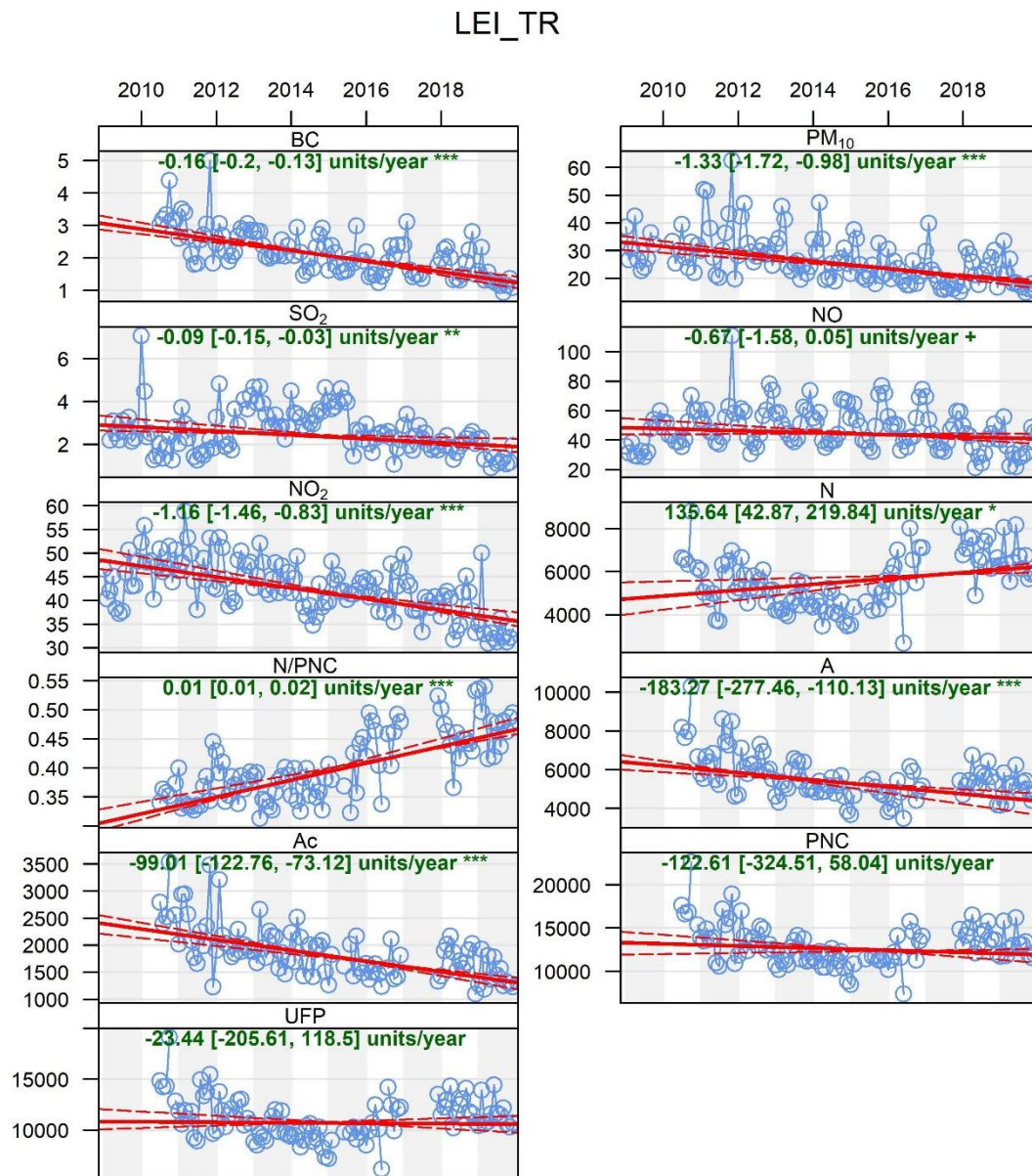


Figure S13. Trend analysis of all pollutants available in Leipzig traffic site (LEI_TR). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

LND_UB

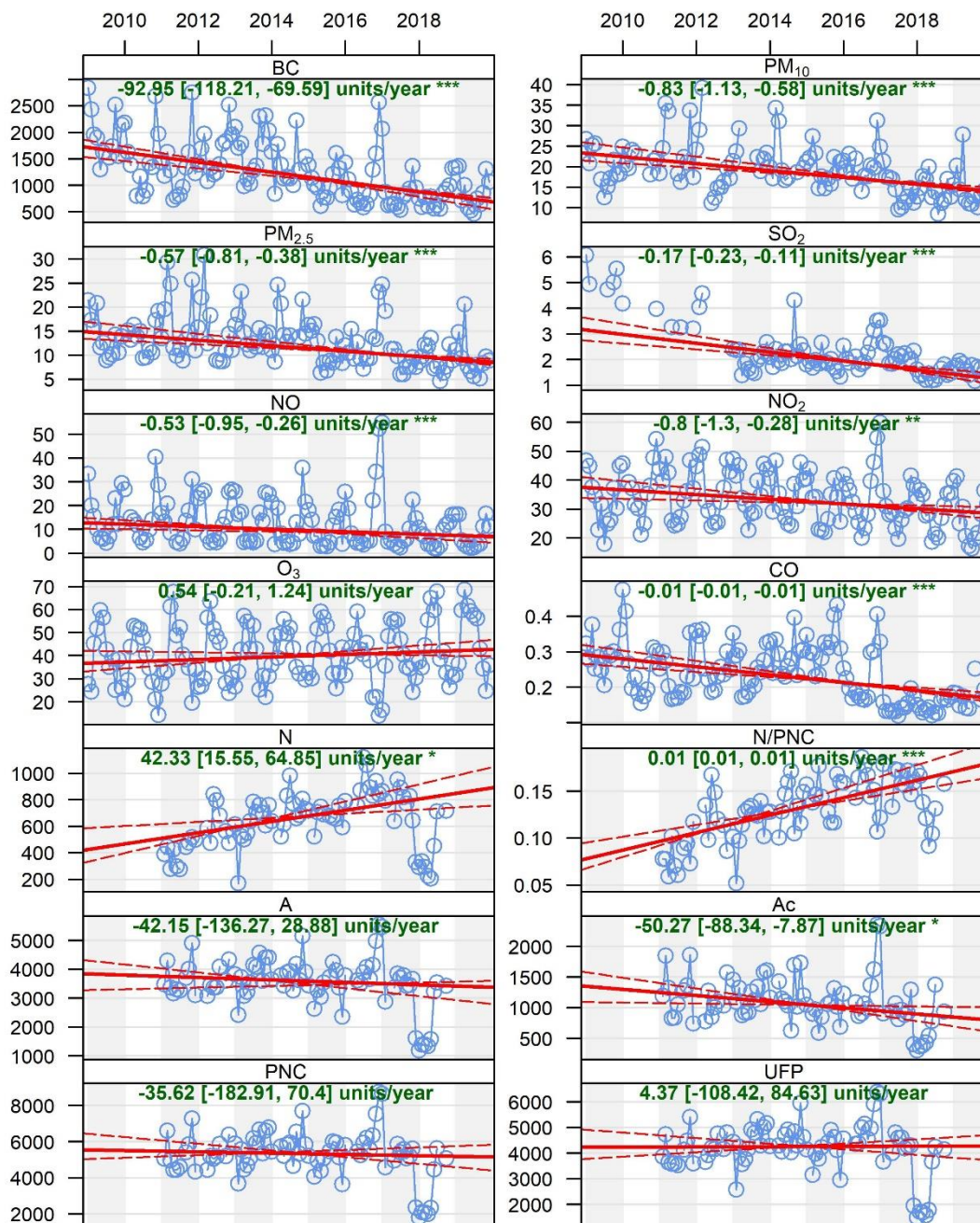


Figure S14. Trend analysis of all pollutants available in London urban background site (LND_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

LND_TR

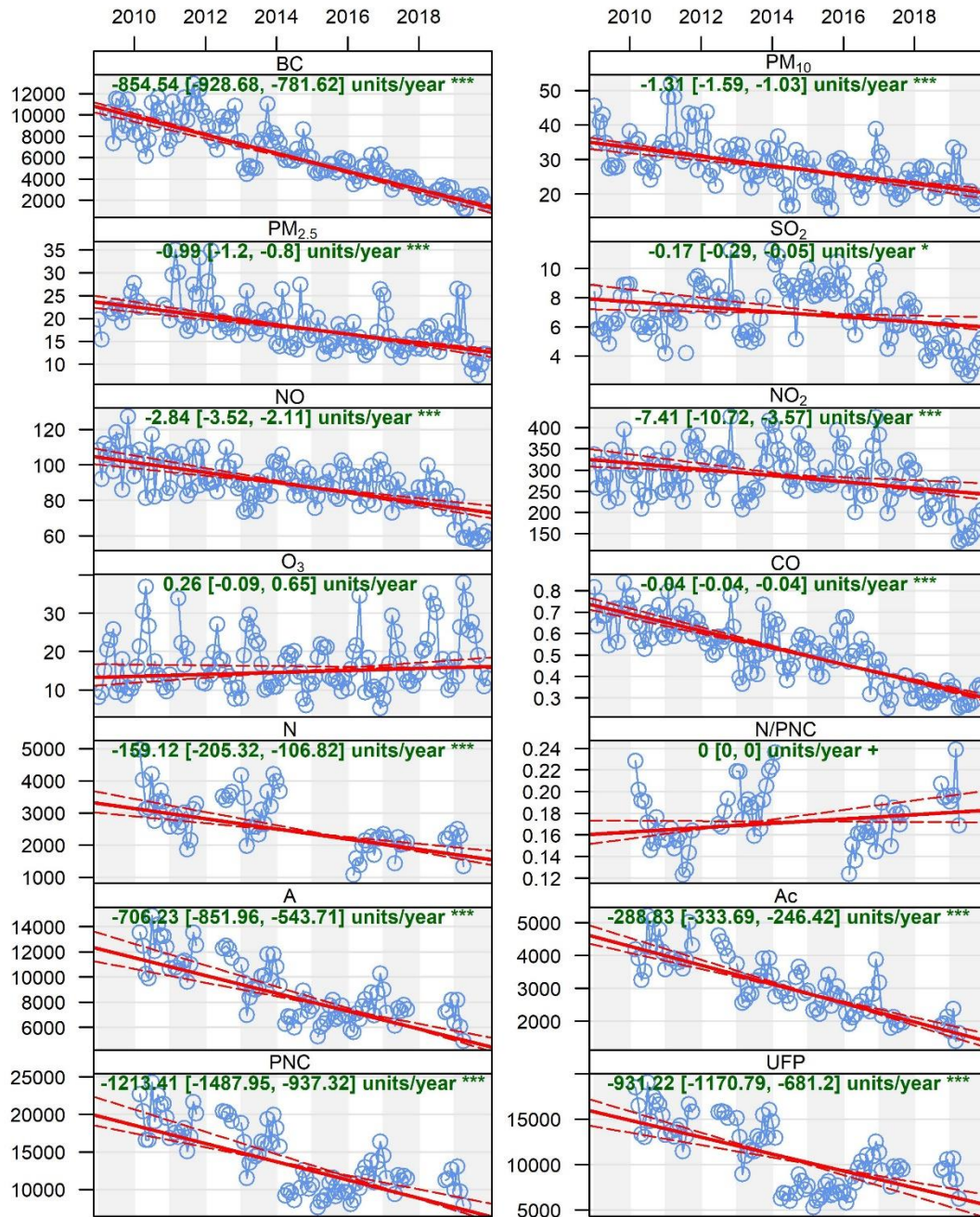


Figure S15. Trend analysis of all pollutants available in London traffic site (LND_TR). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

MAD_UB

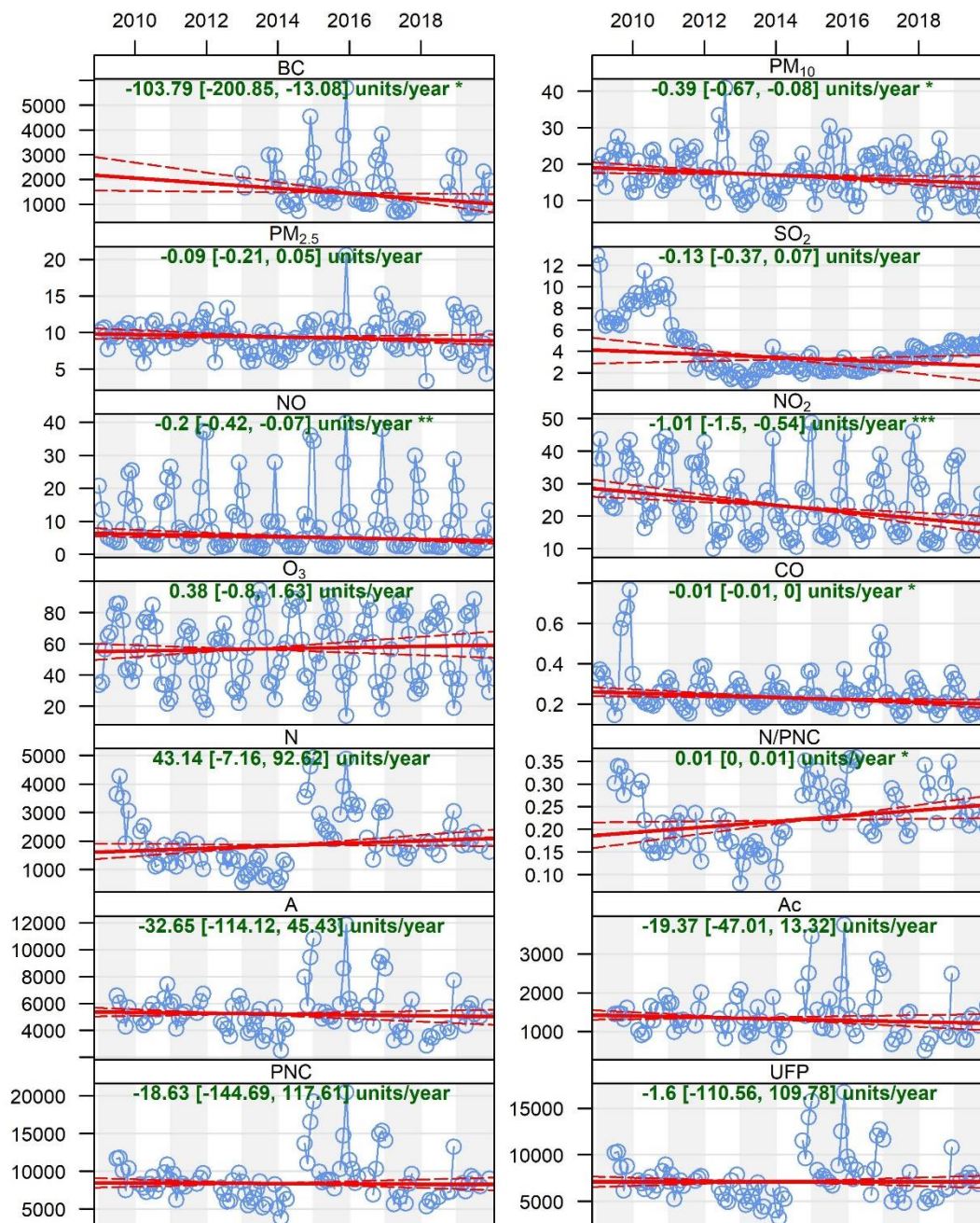


Figure S16. Trend analysis of all pollutants available in Madrid urban background site (MAD_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

MUL_UB

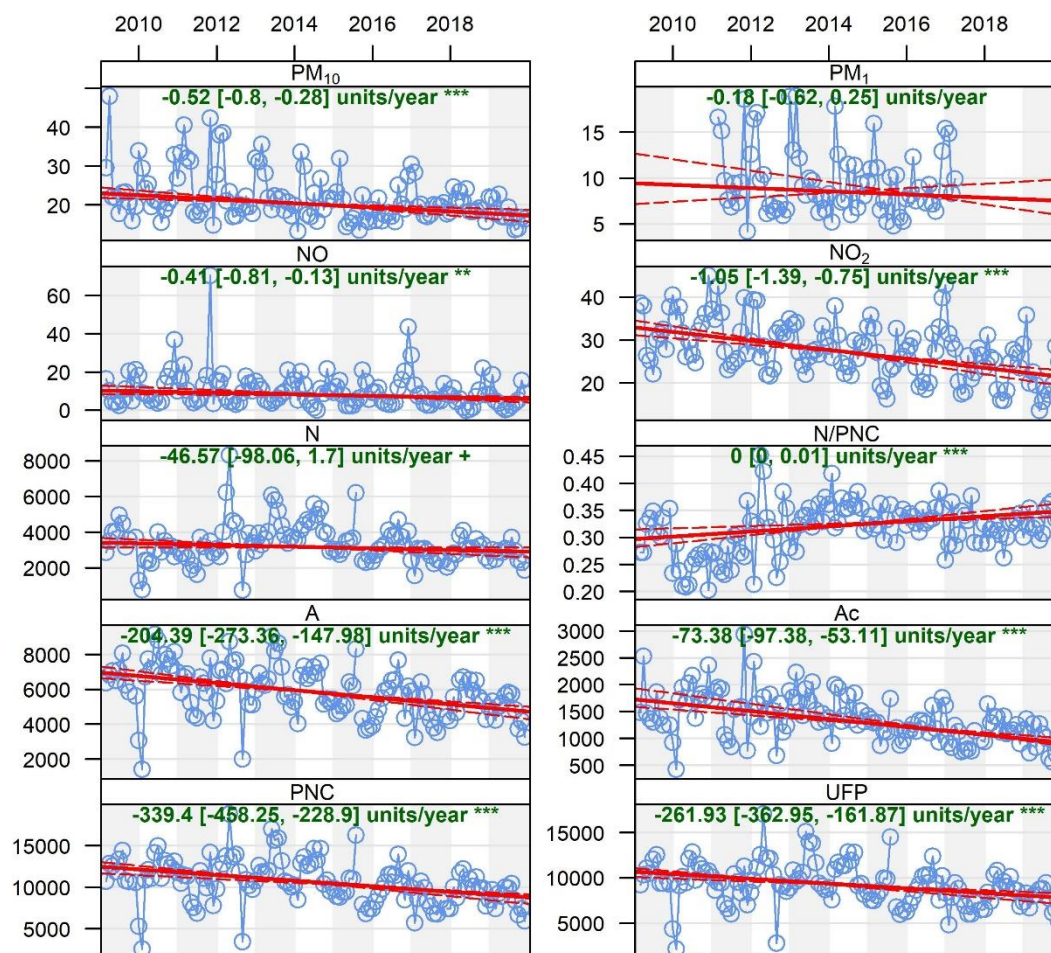


Figure S17. Trend analysis of all pollutants available in Mülheim urban background site (MUL_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

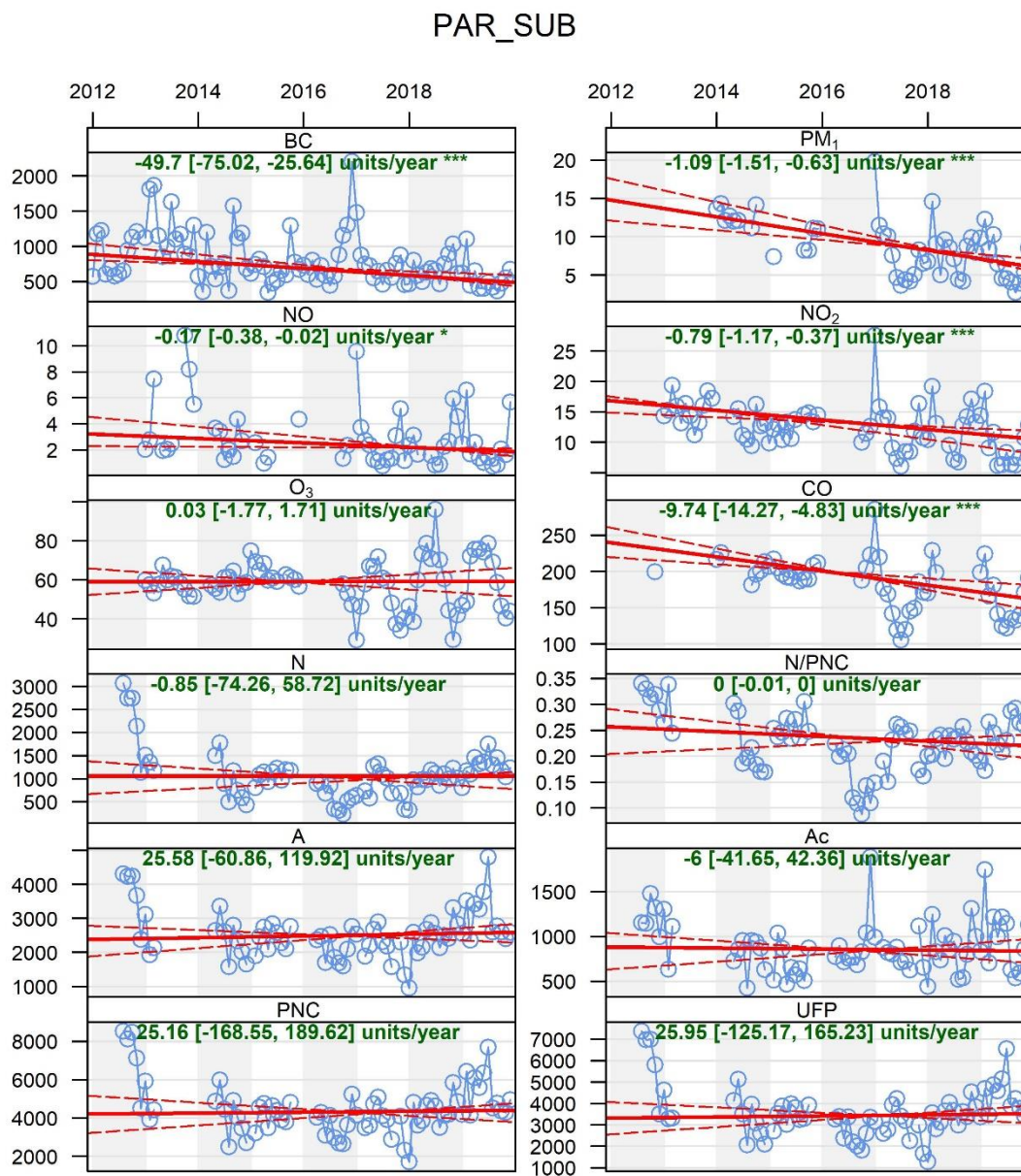


Figure S18. Trend analysis of all pollutants available in Paris suburban background site (PAR_SUB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

PRA_SUB

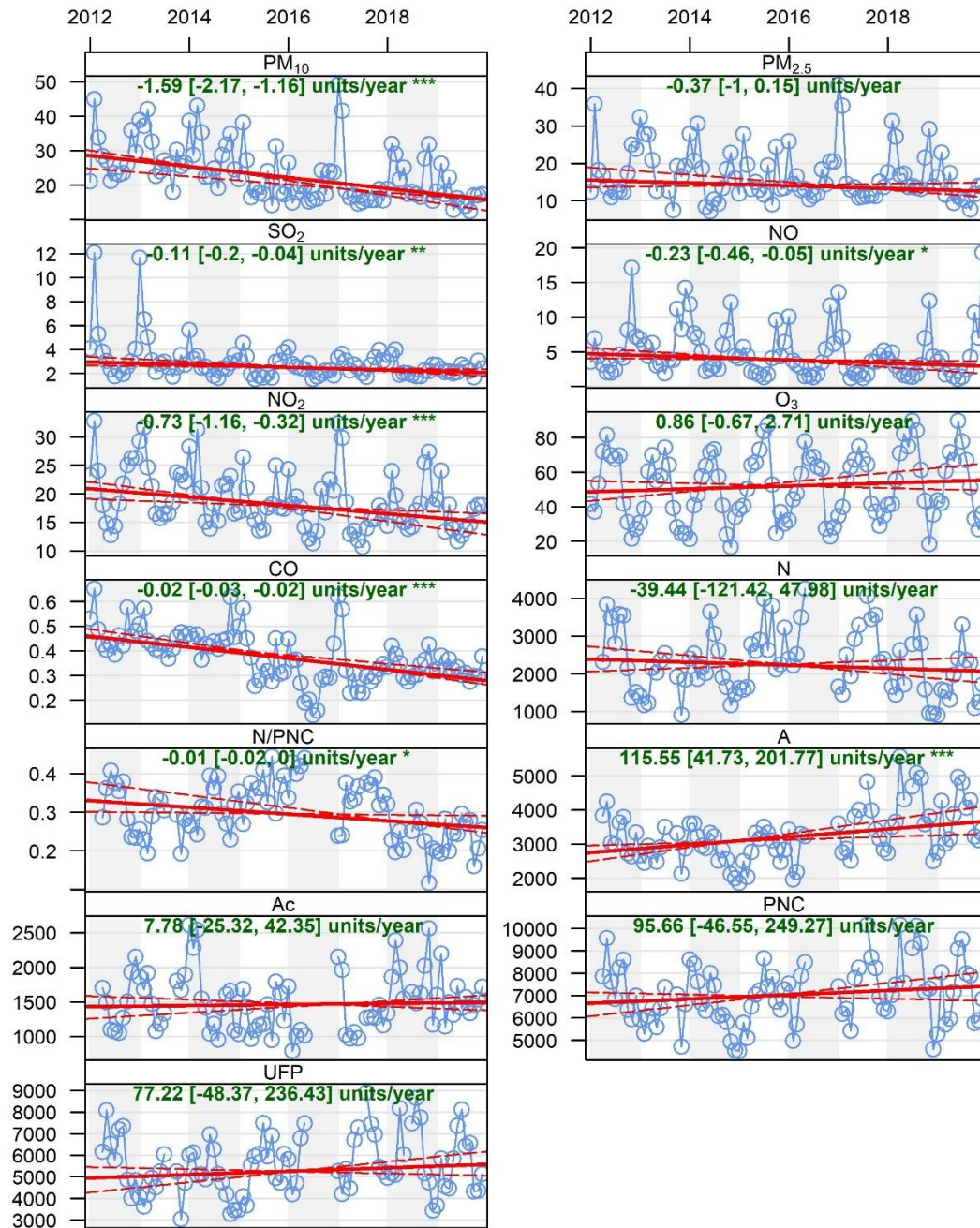


Figure S19. Trend analysis of all pollutants available in Prague suburban background site (PRA_SUB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” (p<0.001), “***” (p<0.01), “**” (p<0.05) and “+” (p<0.1). N, Nucleation; A, Aitken; Ac, Accumulation.

ROC_UB

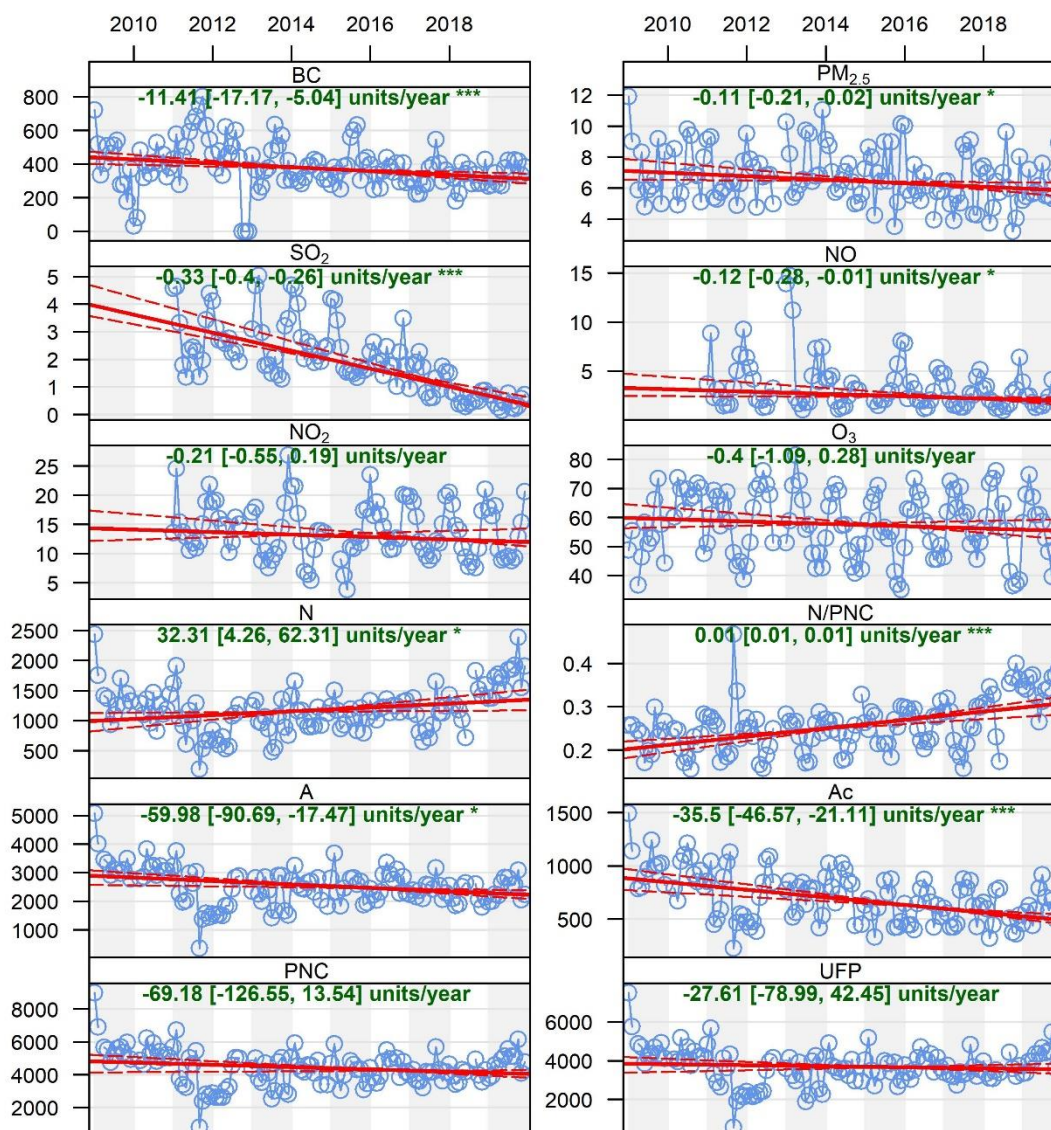


Figure S20. Trend analysis of all pollutants available in Rochester urban background site (ROC_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

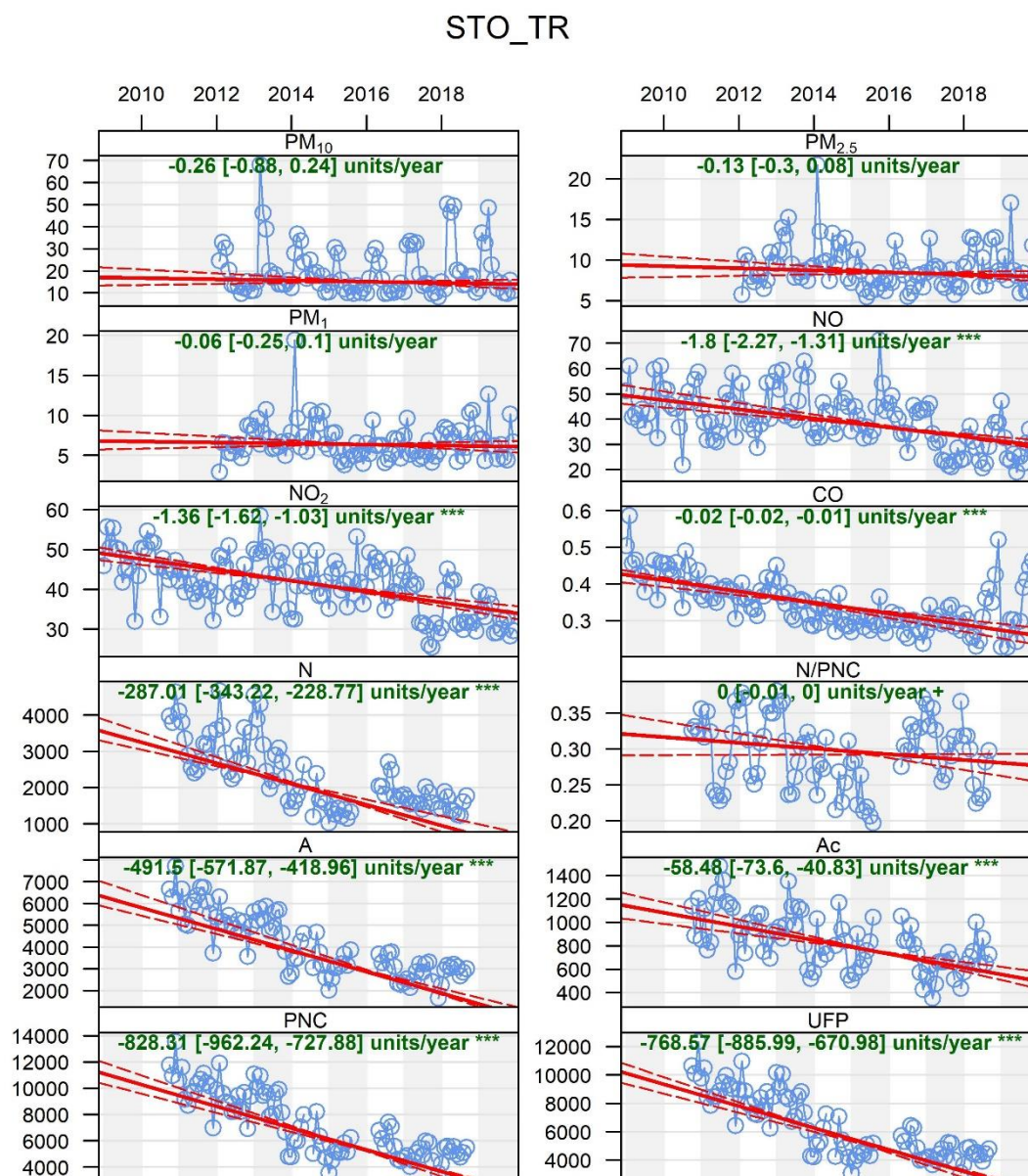


Figure S21. Trend analysis of all pollutants available in Stockholm traffic site (STO_TR). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

ZUR_UB

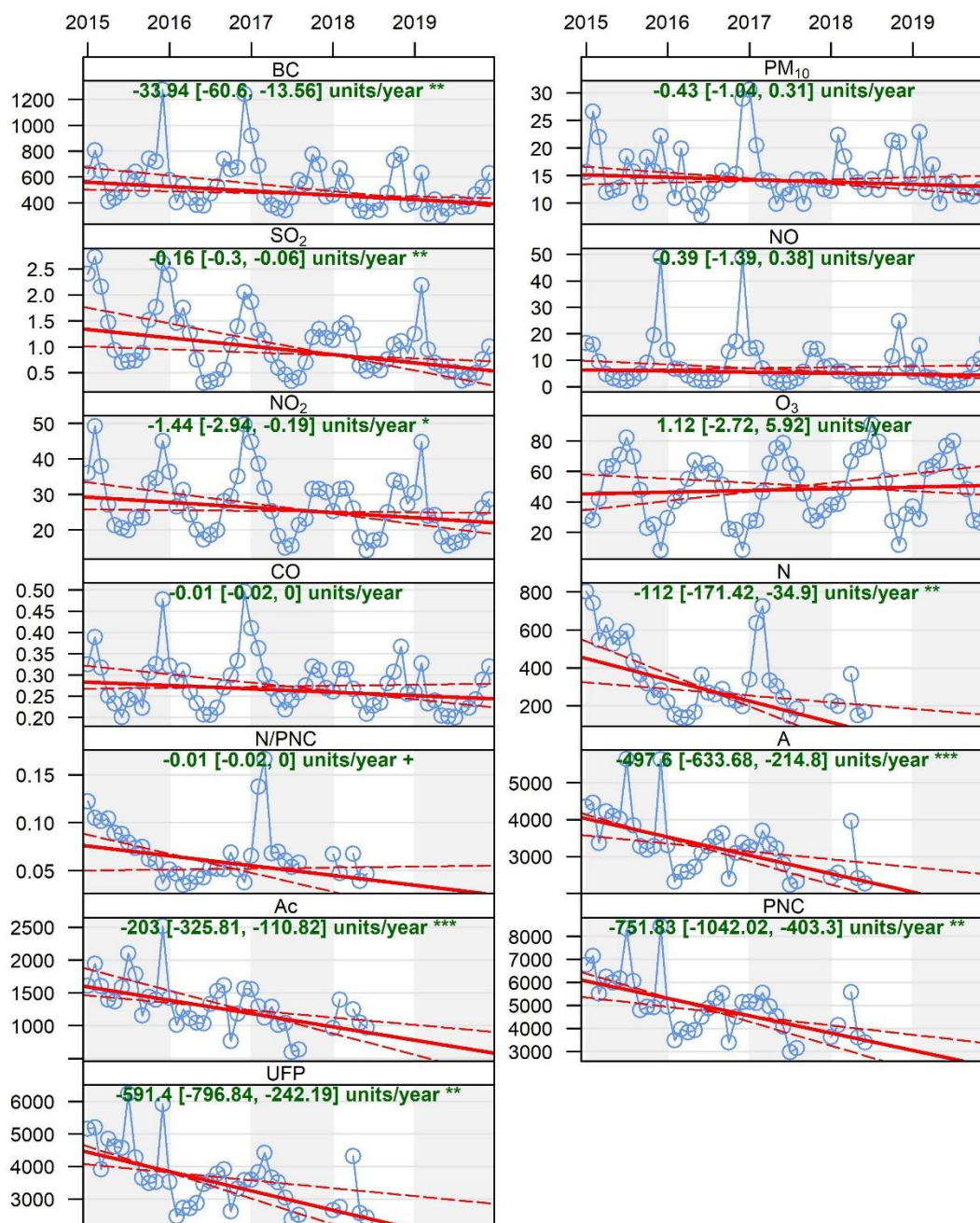


Figure S22. Trend analysis of all pollutants available in Zurich urban background site (ZUR_UB). Trend slopes with 95% confidence interval (CI) were calculated using the Theil-Sen method. Statistical significance is marked by “****” ($p < 0.001$), “***” ($p < 0.01$), “**” ($p < 0.05$) and “+” ($p < 0.1$). N, Nucleation; A, Aitken; Ac, Accumulation.

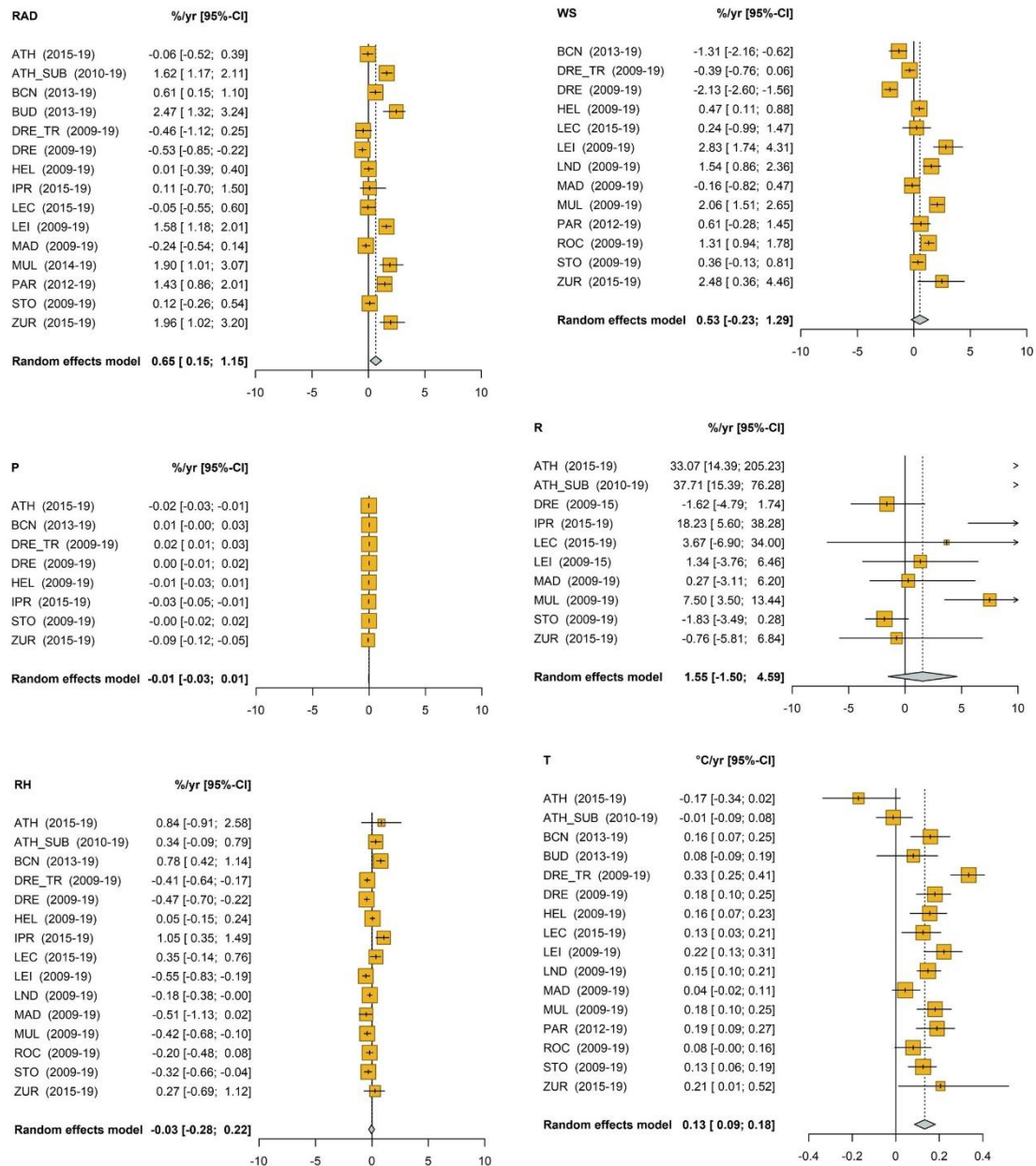


Figure S23. Results of trend analysis and subsequent meta-analysis for six selected meteorological variables in the study cities. The results of solar radiation (RAD), wind speed (WS), atmospheric pressure (P), rain (R) and relative humidity (RH) are reported in % yr⁻¹, with the slope and the 95% confidence intervals. Temperature (T) is reported in °C yr⁻¹.

Supplementary text T1. Analysis of trends by city

Athens (Figures S1 & S2): Both UB and SUB sites showed an increase in NO₂, but decreases in NO, BC, CO and O₃. All PM mass metrics declined as did Aitken and Accumulation modes PNCs. The Nucleation mode decreased at the SUB site due to a long-term gradual reduction in local and regional SO₂ emissions, but increased at the UB site at the shorter period of study coinciding with emissions increasing after the recovery of the financial crisis in Greece. In addition, the introduction of diesel cars in Athens resulted in additional primary NO₂ (National Inventory report of Greece, 2022), when both NO and BC decreased.

Barcelona (Figure S3): There were declines in NO, NO₂, BC, SO₂ and unusually in O₃. None of the particle mass metrics showed any ss trend. Particle number declined in Aitken and Accumulation modes but increased in the Nucleation mode. The increase of the number of cruises might explain this increase (Figure S4, Port de Barcelona, 2023).

Budapest (Figure S5): This site showed declines in NO₂ and NO but an increase in CO. SO₂ and PM₁₀ decreased while O₃ increased, but all three are slight and non-ss. There was a reduction in the Accumulation mode, little change in the Aitken mode and a large increase in the Nucleation one.

Dresden (Figures S6 & S7): There were declining trends in NO, NO₂, SO₂ and BC at the UB site. Ozone showed an increase. PM₁₀ also decreased as did all particle size fraction metrics. This pattern represents an “ideal” behaviour. The TR site showed similar patterns.

Helsinki (Figures S8 & S9): The UB site showed ss declines in NO₂, NO and SO₂. Ozone showed an increase. All of the particle number size modes declined. A similar pattern was seen for the Helsinki TR site, with also decreases in BC and particle mass metrics.

Ispra (Figure S10): At this RB site, the emissions characterised by high SO₂, NO₂ and CO, and low NO/NO₂, were markedly reduced yielding sharp decreasing trends for these pollutants (with a slope 4-10-fold than most of sites). BC and PM₁₀ also decreased, and O₃ increased with a non-ss slope with trends similar to most other sites. PNC and UFP decreased also with similar slopes to other sites. However, the Nucleation mode did not follow a ss trend, although it decreased. Both Aitken and Accumulation modes decreased with ss slopes close to the average of the meta-analysis.

Lecce (Figure S11): Most of the trends at this site were non-ss. A decline is seen in NO₂ and BC, with an increase in NO, O₃ and SO₂. There was a large increase in the Nucleation mode and non-ss decreases in Aitken and Accumulation ones.

Leipzig (Figures S12 & S13): The Leipzig TR site showed declines in NO₂, NO, SO₂ and BC. PM₁₀ also declined, while there was a large increase in the Nucleation mode, and declines in the Aitken and Accumulation ones. At the UB site, there is no gas phase or PM mass data, but the particle size fractions showed very small, non-ss declines in Aitken and Accumulation modes, and a non-ss increase in the Nucleation mode.

London (Figures S14 & S15): The London TR site showed a decrease in NO₂, NO, CO, SO₂ and BC and an increase in O₃. Both PM_{2.5} and PM₁₀ declined, as did all of the three particle size fraction metrics. The London UB site showed similar behaviour for the gaseous pollutants and mass metrics, but a large increase in the Nucleation mode and small decreases in Aitken and Accumulation modes.

Madrid (Figure S16): This UB site followed a typical behaviour, declined in the gaseous primary pollutants, BC and PM mass metrics and increased in O₃. It showed a decline in Aitken and Accumulation modes, but an increase in the Nucleation one.

Mulheim (Figure S17): The UB site showed decreases in NO₂, NO, PM₁ and PM₁₀ and also all three particle size fraction metrics showed declines.

Paris (Figure S18): The SUB site showed ss decreases in NO₂, NO, CO, BC and PM₁, and a non-ss increase in O₃. Nucleation and Accumulation modes decreased but the Aitken one increased, although all three were non-ss.

Prague (Figure S19): The SUB site showed declines in NO₂, NO, CO and SO₂, as well as PM_{2.5} and PM₁₀ mass. Ozone increased, presumably as a result of reduced NO_x. Aitken and Accumulation modes increased while Nucleation decreased but with non-ss.

Rochester, USA (Figure S20): There were small declines in NO₂, NO, SO₂, BC, PM_{2.5} and O₃. There was an increasing trend in the Nucleation mode, but downward trends in Aitken and Accumulation modes.

Stockholm (Figure S21): The TR site showed declining trends in NO₂, NO and CO. PM₁, PM_{2.5} and PM₁₀ declined slightly but the three slopes were non-ss, and the yearly variation in PM₁₀ in this site is strongly weather dependent. The particle size fraction metrics all declined steeply.

Zurich (Figure S22): There were downward trends in BC, PM₁₀ and all gaseous pollutants except O₃. All particle size fraction metrics showed substantial rates of decline.

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Port de Barcelona, 2023. Estadísticas del Puerto de Barcelona. <https://www.portdebarcelona.cat/es/web/autoritat-portuaria/estadisticas>

Supplement of paper III

Source apportionment of ultrafine particles in urban Europe

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Table S1. Coordinates of the sites, instrumentation used to measure UFP-PNSD and number of hours of data available for the source apportionment analysis. Coord., Coordinates; Alt., Altitude; SMPS, Scanning Mobility Particle Sizer; CPC, Condensation Particle Counter; MAAP, Multi-Angle Absorption Photometer; DMPS, Differential Mobility Particle Sizer; TSMPS, Twin Scanning Mobility Particle Sizer; DMA, Differential Mobility Analyser; TDMPS, Twin Differential Mobility Particle Sizer; UCPC, Ultrafine Condensation Particle Counter; OPC, Optical Particle Counter.

Station	Coord.; (Alt., m a.s.l.)	UFP-PNSD instrumentation	Hours
ANT_UB	51.21 N, 4.43 E; (10)	SMPS Grimm C5420 long DMA	16970
ATH_UB	38.00 N, 23.72 E; (110)	SMPS TSI 3034	14130
BCN_UB	41.39 N, 2.13E; (80)	SMPS TSI 3080 + CPC TSI 3772	38941
BUD_UB	47.48 N, 19.06 E; (115)	DMPS + CPC TSI 3775	40481
DRE_UB	51.04 N, 13.73 E; (120)	TROPOS-SMPS uses Vienna-type DMA 28 cm + CPC TSI 3772	57375
GRA_UB	37.16 N, 3.61 W; (680)	SMPS TSI 3082 + CPC TSI 3772	10346
HEL_UB	60.12 N, 24.58 E; (26)	TDMPS Hauke-type DMA 10.9 cm + CPC TSI 3025/3756 and Hauke-type DMA 28 cm + CPC 3010/3772	71072
LEC_UB	40.20 N, 18.07 E; (50)	TROPOS-SMPS uses a Vienna-type DMA 28 cm + CPC TSI 3772	24376
LEI_UB	51.35 N, 12.43 E; (113)	TROPOS-TDMPS uses Vienna-type DMAs 11 and 28 cm + CPC TSI 3025/3010	81903
LND_UB	51.52 N, 0.21 W; (27)	SMPS TSI 3080 + CPC TSI 3775 with long DMA	37249
MAD_UB	40.45 N, 3.73 W; (669)	SMPS TSI 3080L + CPC TSI 3775	20424
MUL_UB	51.45 N, 6.87 E; (39)	SMPS TSI 3080 + CPC TSI 3772	66404
ROC_UB	43.15 N, 77.55 W; (137)	SMPS TSI 3071 + CPC TSI 3010	62972
ZUR_UB	47.38 N, 8.53 E; (410)	SMPS TSI 3034 + Nafion aerosol dryer	23809
DRE_TR	51.07 N, 13.74 E; (116)	TROPOS-TSMPS uses Vienna-type DMAs 11 and 28 cm + CPC TSI 3025/3010	69697
HEL_TR	60.19 N, 24.95 E; (26)	UHEL DMPS Vienna-type DMA + CPC Airmodus A20	32085
LEI_TR	51.34 N, 12.38 E; (111)	TROPOS-TDMPS uses Vienna-type DMAs 11 and 28 cm + CPC TSI 3025/3010	61046
LND_TR	51.52 N, 0.15 W; (35)	SMPS TSI 3080 + CPC TSI 3775 with long DMA	26653
STO_TR	59.32 N, 18.05 E; (20)	DMPS Stockholm university handmade + CPC TSI 3010/3775	42333
ATH_SUB	37.99 N, 23.82 E; (270)	SMPS TSI 3080 + CPC TSI 3772	38180
LIL_SUB	50.61 N, 3.14 E; (70)	SMPS TSI 3082 + CPC TSI 3750	11049
PAR_SUB	48.71 N, 2.16 E; (162)	SMPS GRIMM 5416 + OPC GRIMM	23552
PRA_SUB	50.13 N, 14.38 E; (277)	SMPS TSI 3034 rebuilt at TROPOS	42464
IPR_RB	45.80 N, 8.63 E; (209)	DMPS Vienna-type, home-made + CPC TSI 3010/3772	20819

Table S2. Selection of the parameters C_3 and K optimising PMF solutions.

Station	C_3	K
ANT_UB	0.05	0.08
ATH_UB	0.03	0.03
BCN_UB	0.01	0.04
BUD_UB	0.1	0.05
DRE_UB	0.03	0.06
GRA_UB	0.03	0.05
HEL_UB	0.07	0.09
LEC_UB	0.07	0.06
LEI_UB	0.04	0.05
LND_UB	0.06	0.06
MAD_UB	0.05	0.05
MUL_UB	0.03	0.05
ROC_UB	0.09	0.09
ZUR_UB	0.05	0.05
DRE_TR	0.05	0.06
HEL_TR	0.1	0.09
LEI_TR	0.06	0.08
LND_TR	0.001	0.06
STO_TR	0.1	0.07
ATH_SUB	0.07	0.05
LIL_SUB	0.05	0.05
PAR_SUB	1.5	0.08
PRA_SUB	0.06	0.07
IPR_RB	0.08	0.07

Table S3. Splitting of the factors and proxies used for traffic emissions for each site, and Photonucleation contribution initially (before the split and summation of the two Photonucleation).

Station	Proxy	Splitting of the factors	% Photonucleation before split
ANT_UB	NO _x	nucleation into Traffic-Nucleation + Photonucleation	
ATH_UB	NO _x	traffic into Traffic-1 + Photonucleation	7%
BCN_UB	BC	traffic into Traffic-1 + Photonucleation	24%
BUD_UB	NO	nucleation into Traffic-Nucleation + Photonucleation	
DRE_UB	NO ₂	traffic into Traffic-1 + Photonucleation	19%
GRA_UB	BC	nucleation into Traffic-Nucleation + Photonucleation	
LEC_UB	NO _x	nucleation into Traffic-Nucleation + Photonucleation	
LEI_UB	NO ₂	splitting of both nucleation and traffic factors	16%
MAD_UB	BC	nucleation into Traffic-Nucleation + Photonucleation	
MUL_UB	NO ₂	nucleation into Traffic-Nucleation + Photonucleation	
ROC_UB	NO ₂	nucleation into Traffic-Nucleation + Photonucleation	
DRE_TR	NO _x	nucleation into Traffic-Nucleation + Photonucleation	
LEI_TR	NO _x	nucleation into Traffic-Nucleation + Photonucleation	
ATH_SUB	NO ₂	traffic into Traffic-1 + Photonucleation	15%
LIL_SUB	NO ₂	nucleation into Traffic-Nucleation + Photonucleation	

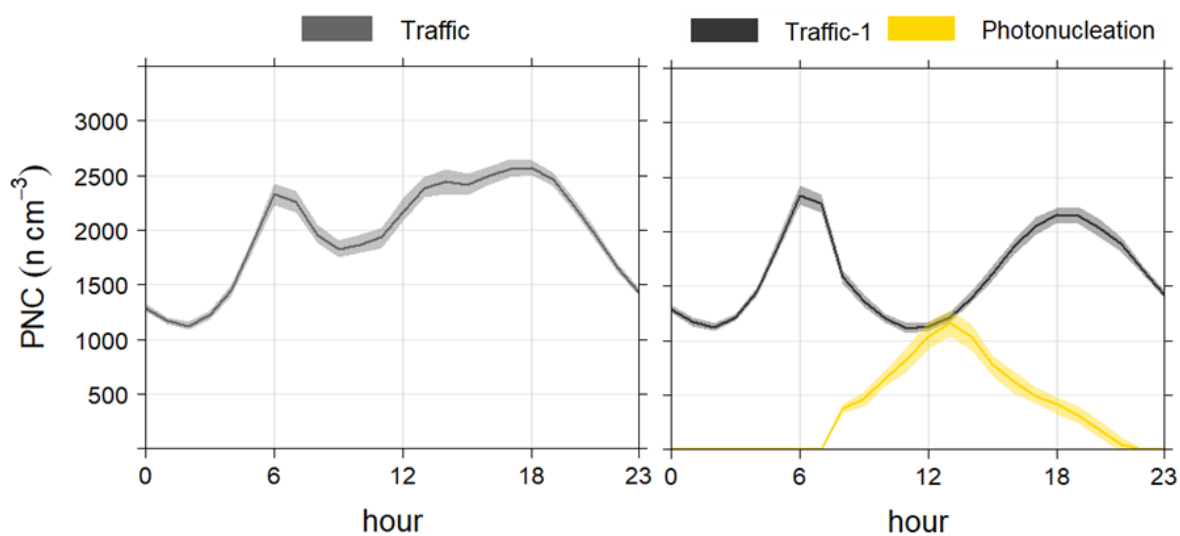


Figure S1. Splitting of traffic into *Traffic-1* and *Photonucleation* for DRE_UB using NO₂ as a proxy for traffic emissions.

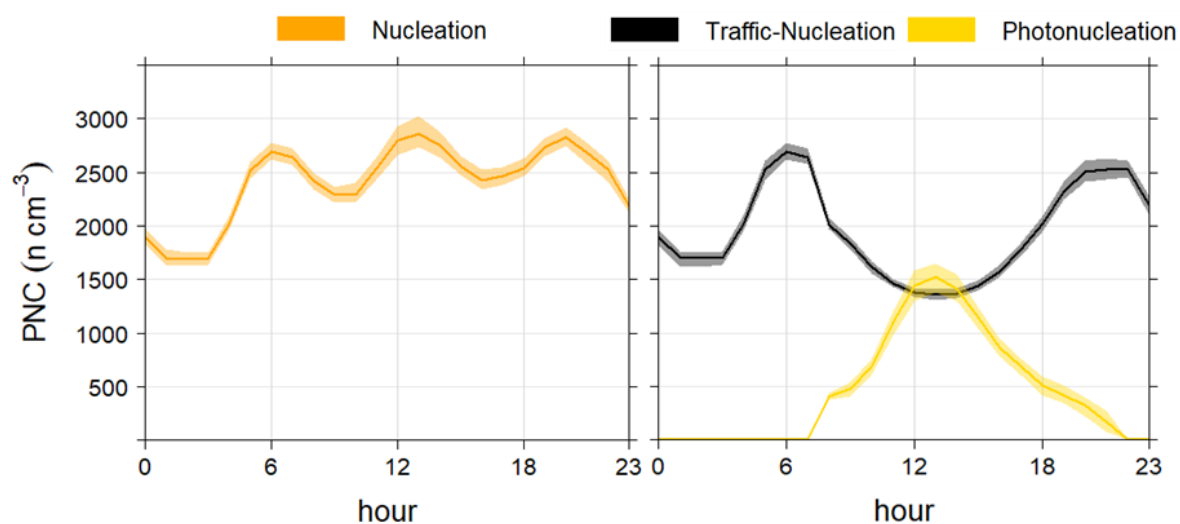


Figure S2. Splitting of nucleation into *Photonucleation* and *Traffic-Nucleation* for MUL_UB using NO₂ as a proxy for traffic emissions.

Supplementary text T1. Source apportionment by site

For each site it is presented the results of the source apportionment as follows: Profiles for particle number size distribution (PNSD); contributions to the variance of particulate matter (PM), black carbon (BC) and gaseous pollutants (SO₂, NO, NO₂, O₃ and CO); Daily, monthly and weekly patterns of particle number concentrations (PNCs) of each source; and polar plots indicating the wind speeds and directions associated with higher concentrations of each source. A description of the sources can be found in the manuscript.

ANT_UB (Figures S3-S5 and Table S4): Six sources were identified in the UB site in Antwerp (Regional-1, Photonucleation, Traffic-1, Regional-2, Traffic-2 and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

ATH_UB (Figures S6-S8 and Table S5): Five sources were identified in the UB site in Athens (Photonucleation, Domestic heating, Urban background, Traffic-1 and Mixed traffic). The factor Traffic-1 was split into two factors due to the presence of peaks at rush hours and midday (Traffic-1 and Photonucleation). Therefore, both Photonucleation factors were summed.

BCN_UB (Figures S9-S14 and Table S6): Six sources were identified in the UB site in Barcelona (Regional-2, Photonucleation, Traffic-1, Long-distance transport, Traffic-2 and Mixed traffic). The factor Traffic-1 was split into two factors due to the presence of peaks at rush hours and midday (Traffic-1 and Photonucleation). Therefore, both Photonucleation factors were summed.

BUD_UB (Figures S15-S17 and Table S7): Six sources were identified in the UB site in Budapest (Regional-2, Mixed traffic, Urban background, Regional-1, Photonucleation and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

DRE_UB (Figures S18-S20 and Table S8): Six sources were identified in the UB site in Dresden (Regional-2, Traffic-1, Traffic-2, Photonucleation, Mixed traffic and Regional-1). The factor Traffic-1 was split into two factors due to the presence of peaks at rush hours and midday (Traffic-1 and Photonucleation). Therefore, both Photonucleation factors were summed.

GRA_UB (Figures S21-S23 and Table S9): Six sources were identified in the UB site in Granada (Mixed traffic, Traffic-2, Traffic-1, Photonucleation, Regional-1 and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

HEL_UB (Figures S24-S26 and Table S10): Four sources were identified in the UB site in Helsinki (Mixed traffic, Regional-2, Traffic-Nucleation and Regional-1).

LEC_UB (Figures S27-S29 and Table S11): Seven sources were identified in the UB site in Lecce (Regional-1, Traffic-2, Traffic-1, Domestic heating, Regional-2, Photonucleation and Traffic-

Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

LEI_UB (Figures S30-S32 and Table S12): Seven sources were identified in the UB site in Leipzig (Photonucleation, Mixed traffic, Regional-2, Regional-1, Traffic-2, Traffic-1 and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday. Also, the factor Traffic-1 was split into two factors (Traffic-1 and Photonucleation). Therefore, both Photonucleation factors were summed.

LND_UB (Figures S33-S35 and Table S13): Four sources were identified in the UB site in London (Traffic-2, Traffic-1, Regional-1 and Photonucleation).

MAD_UB (Figures S36-S38 and Table S14): Six sources were identified in the UB site in Madrid (Traffic-2, Traffic-1, Regional-2, Urban background, Photonucleation and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

MUL_UB (Figures S39-S41 and Table S15): Seven sources were identified in the UB site in Mulheim (Mixed traffic, Photonucleation, Traffic-1, Regional-1, Urban background, Traffic-2 and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

ROC_UB (Figures S42-S44 and Table S16): Seven sources were identified in the UB site in Rochester (Regional-2, Traffic-2, Traffic-1, Regional-1, Domestic heating, Photonucleation and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

ZUR_UB (Figures S45-S47 and Table S17): Six sources were identified in the UB site in Rochester (Photonucleation, Regional-2, Traffic-1, Traffic-2, Domestic heating and Regional-1).

DRE_TR (Figures S48-S50 and Table S18): Six sources were identified in the TR site in Dresden (Traffic-2, Photonucleation, Traffic-1, Regional-2, Regional-1 and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

HEL_TR (Figures S51-S53 and Table S19): Four sources were identified in the TR site in Helsinki (Regional-2, Regional-1, Traffic-Nucleation and Mixed traffic).

LEI_TR (Figures S54-S56 and Table S20): Five sources were identified in the TR site in Leipzig (Traffic-2, Regional-1, Photonucleation, Traffic-1 and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

LND_TR (Figures S57-S59 and Table S21): Four sources were identified in the TR site in London (Traffic-1, Traffic-2, Traffic-Nucleation and Regional-1).

STO_TR (Figures S60-S62 and Table S22): Four sources were identified in the TR site in Stockholm (Traffic-Nucleation, Traffic-2, Traffic-1, and Regional-1).

ATH_SUB (Figures S63-S65 and Table S23): Four sources were identified in the SUB site in Athens (Traffic-2, Regional-2, Photonucleation and Traffic-1). The factor Traffic-1 was split into two factors due to the presence of peaks at rush hours and midday (Traffic-1 and Photonucleation). Therefore, both Photonucleation factors were summed.

LIL_SUB (Figures S66-S68 and Table S24): Six sources were identified in the SUB site in Lille (Urban background, Photonucleation, Regional-1, Regional-2, Mixed traffic and Traffic-Nucleation). Traffic-Nucleation and Photonucleation were obtained after splitting a Nucleation factor due to the presence of peaks at rush hours and midday.

PAR_SUB (Figures S69-S71 and Table S25): Four sources were identified in the SUB site in Paris (Traffic-1, Regional-1, Regional-2 and Traffic-2).

PRA_SUB (Figures S72-S74 and Table S26): Four sources were identified in the SUB site in Prague (Mixed traffic, Domestic heating, Photonucleation and Regional-1).

IPR_RB (Figures S75-S77 and Table S27): Five sources were identified in the RB site in Ispra (Mixed traffic, Domestic heating, Regional-1, Photonucleation and Regional-2).

Table S4. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in ANT_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃
1	271 & 30	14	22	35	2	5	9	0
2	11	18	14	7	15	20	23	0
3	39	8	12	7	24	0	19	5
4	10 & 191	4	18	15	9	0	3	61
5	83	29	13	12	15	7	26	0
UNDETERMINED		27	21	24	35	68	19	34

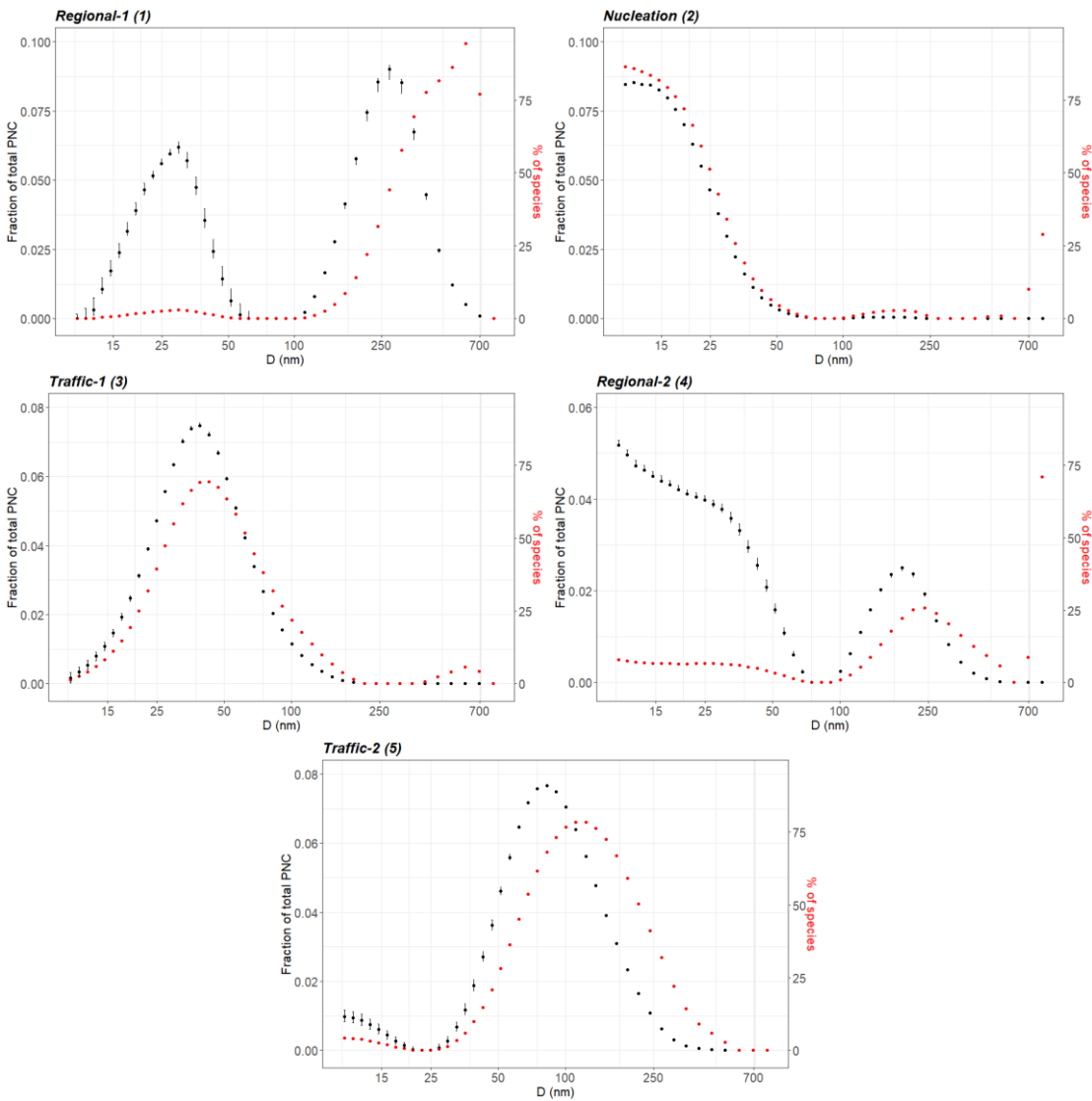


Figure S3. Particle number size distributions of each factor identified by PMF (before splitting) in ANT_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

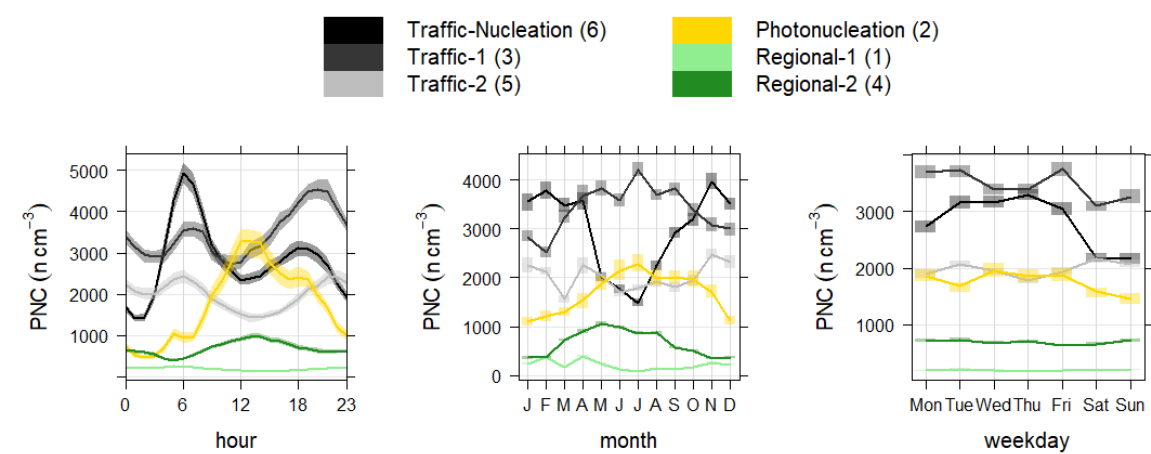


Figure S4. Daily, monthly and weekly patterns of the sources identified (after splitting) in ANT_UB.

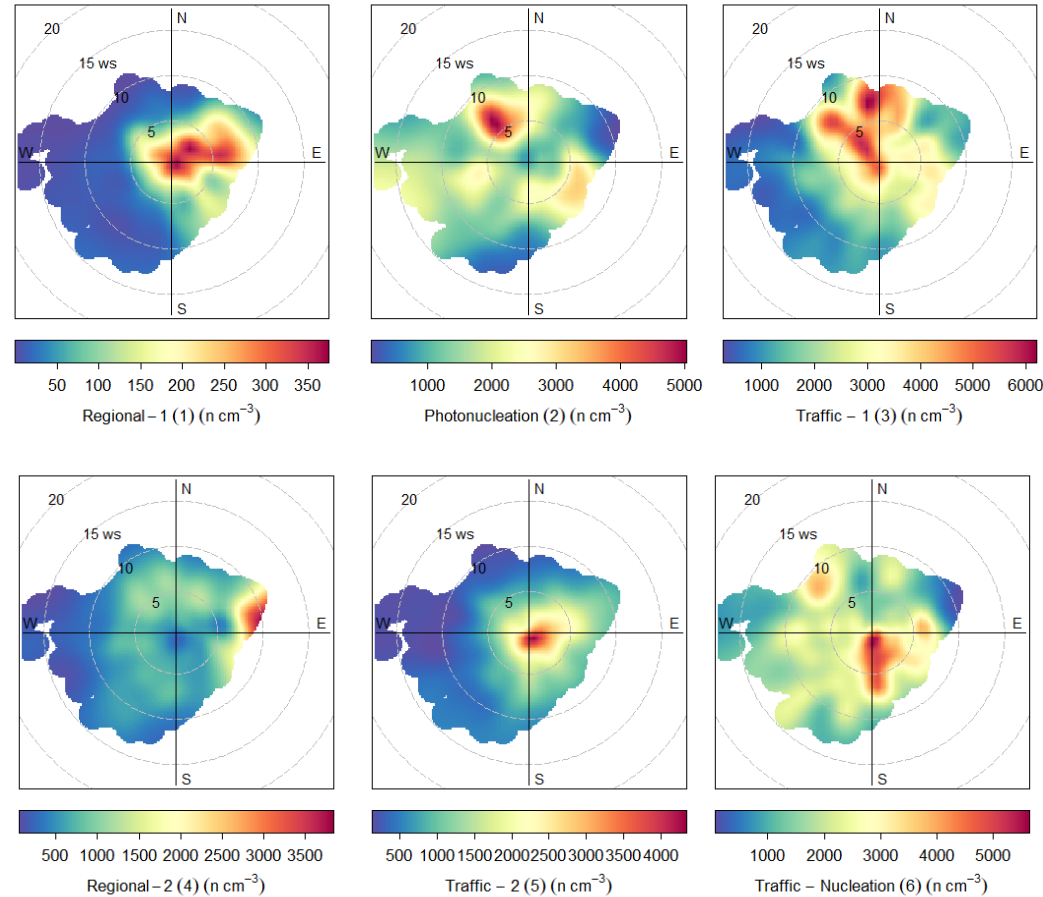


Figure S5. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in ANT_UB.

Table S5. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in ATH_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	NO	NO ₂	O ₃	CO
1	16 (60)	1	5	2	4	41	3
2	84	33	15	18	11	0	27
3	160 (28)	28	37	3	22	0	30
4	28	7	3	5	12	0	13
5	47	9	13	3	30	0	16
UNDETERMINED		23	27	69	20	59	11

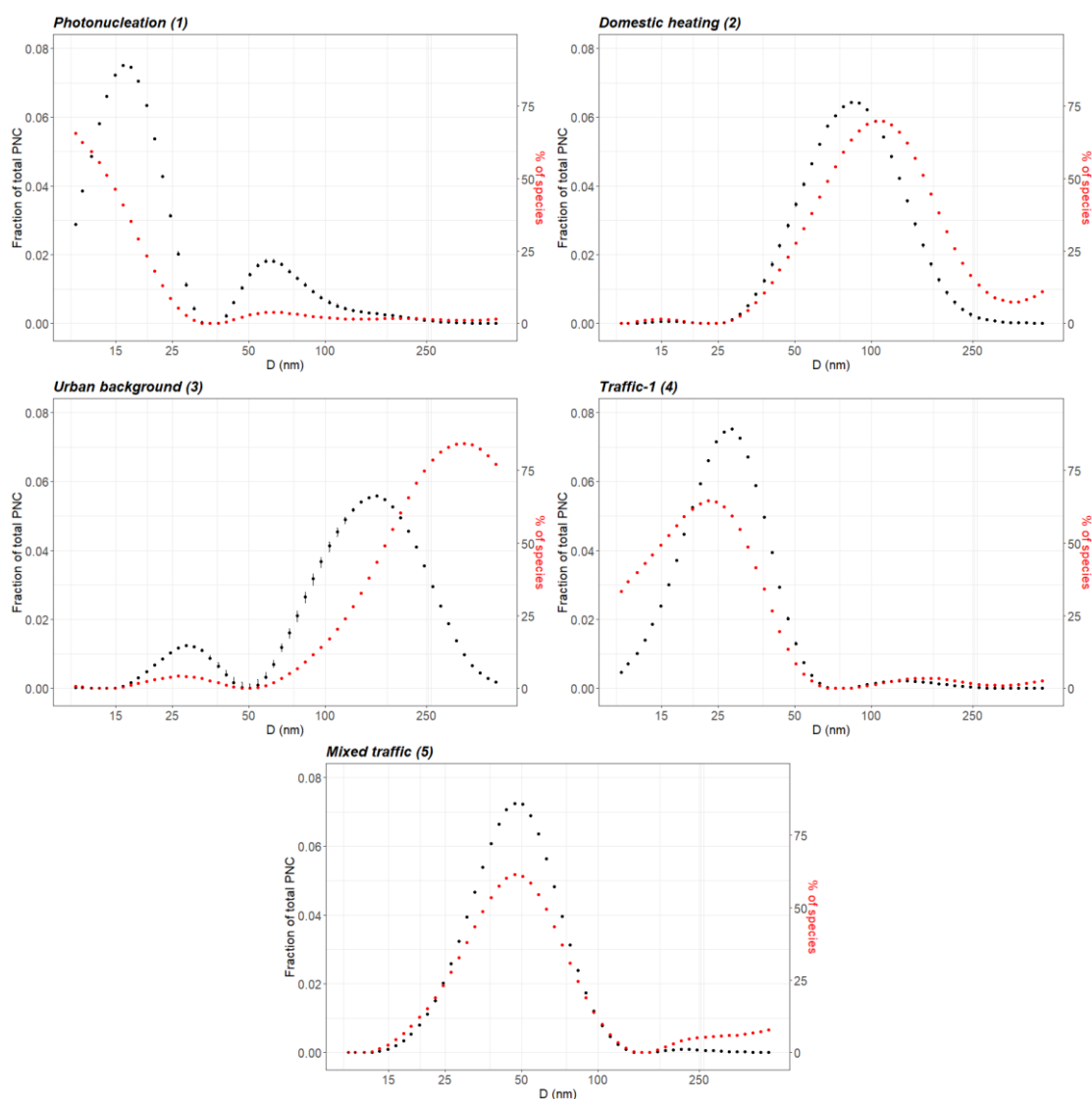


Figure S6. Particle number size distributions of each factor identified by PMF (before splitting) in ATH_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

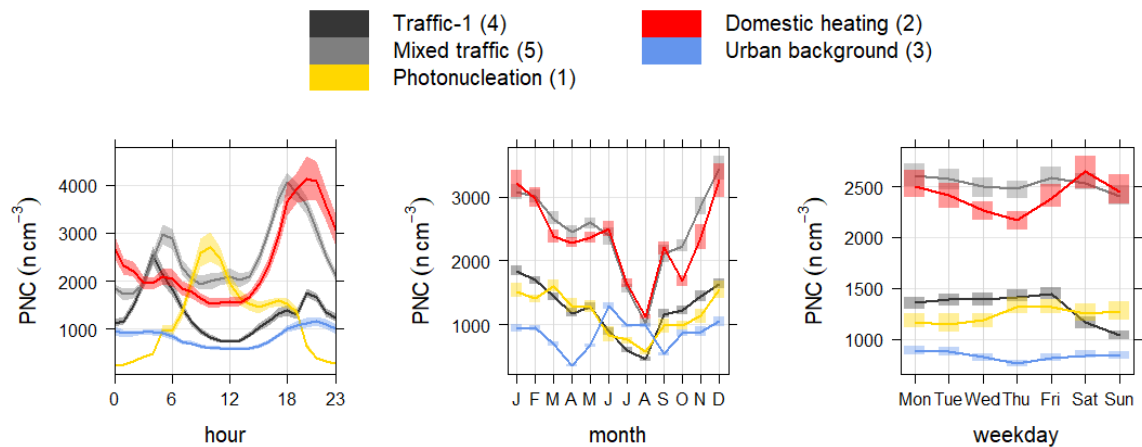


Figure S7. Daily, monthly and weekly patterns of the sources identified (after splitting) in ATH_UB.

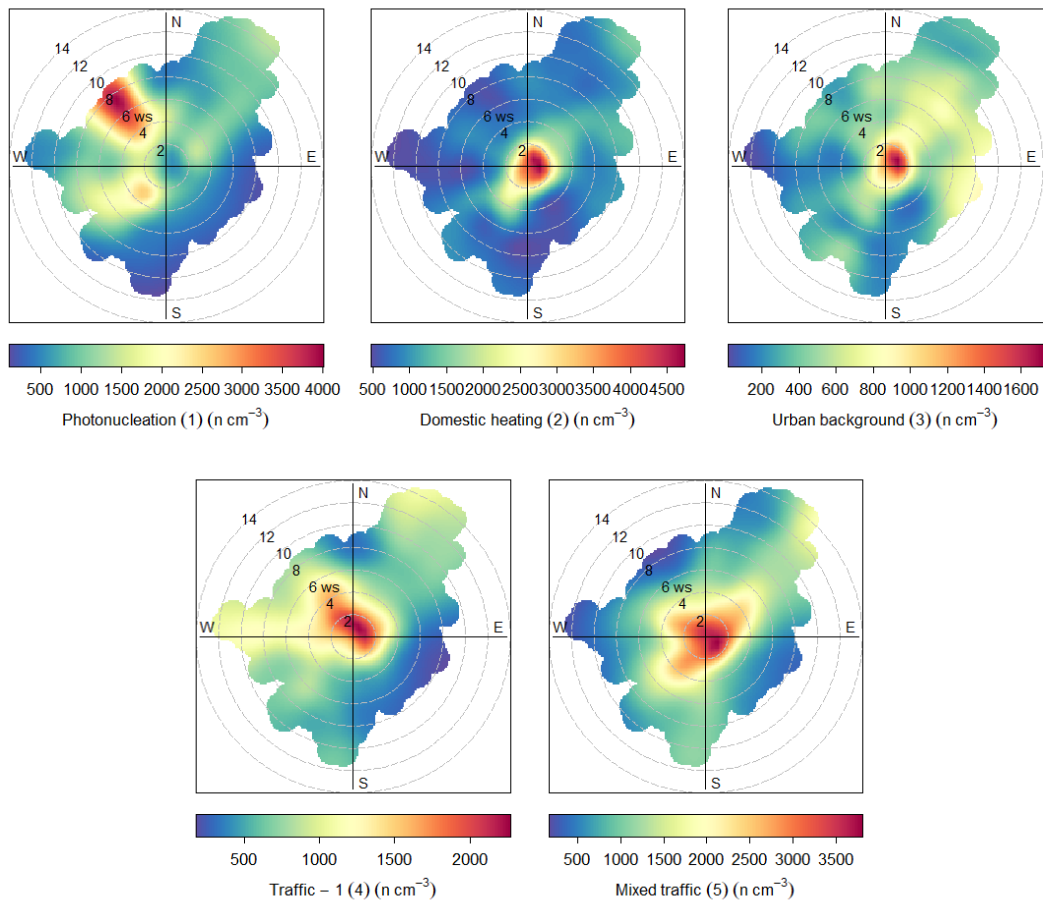


Figure S8. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s⁻¹) in ATH_UB.

Table S6. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in BCN_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃	CO
1	151 (31)	13	33	37	26	3	8	18	22
2	13	0	6	4	17	2	2	25	7
3	27	7	3	1	0	5	7	0	8
4	300 (69, 13)	7	16	18	0	1	3	0	0
5	88 (18)	42	14	15	12	9	40	0	16
6	43	12	10	9	16	6	28	0	26
UNDETERMINED		18	18	16	27	74	12	57	22

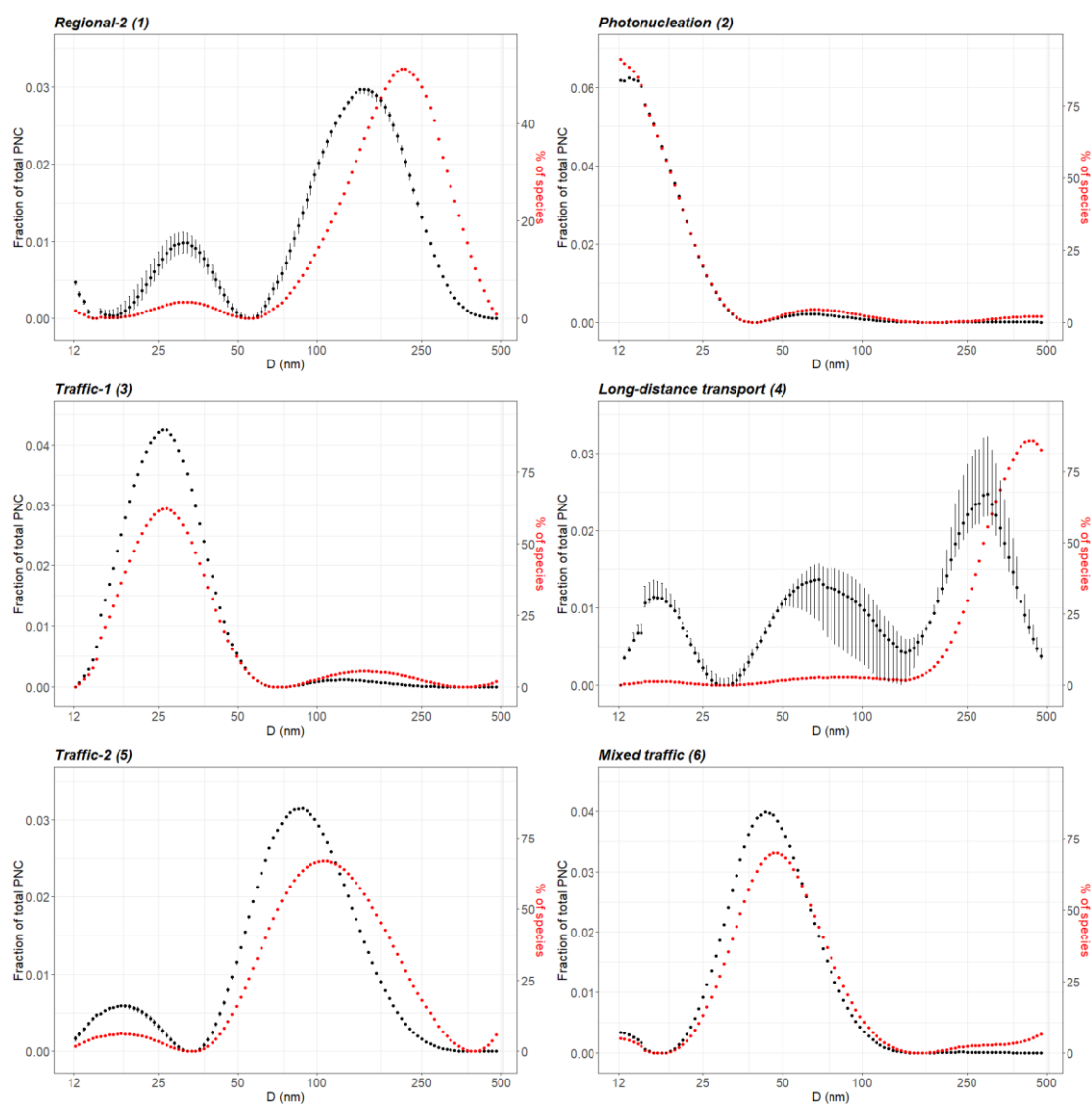


Figure S9. Particle number size distributions of each factor identified by PMF (before splitting) in BCN_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

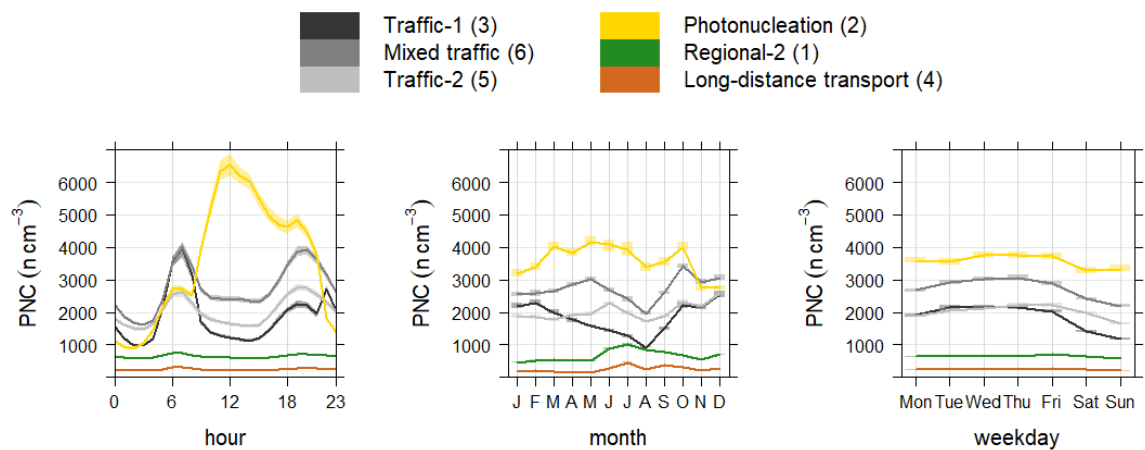


Figure S10. Daily, monthly and weekly patterns of the sources identified (after splitting) in BCN_UB.

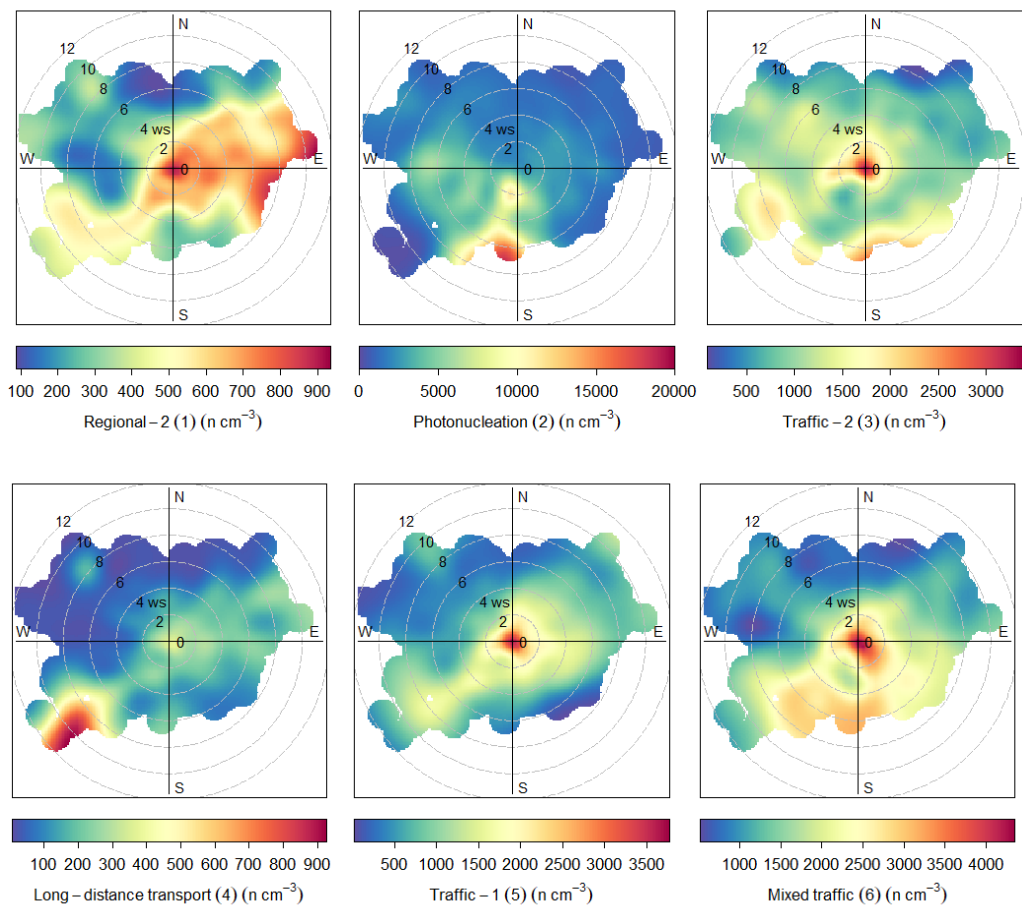
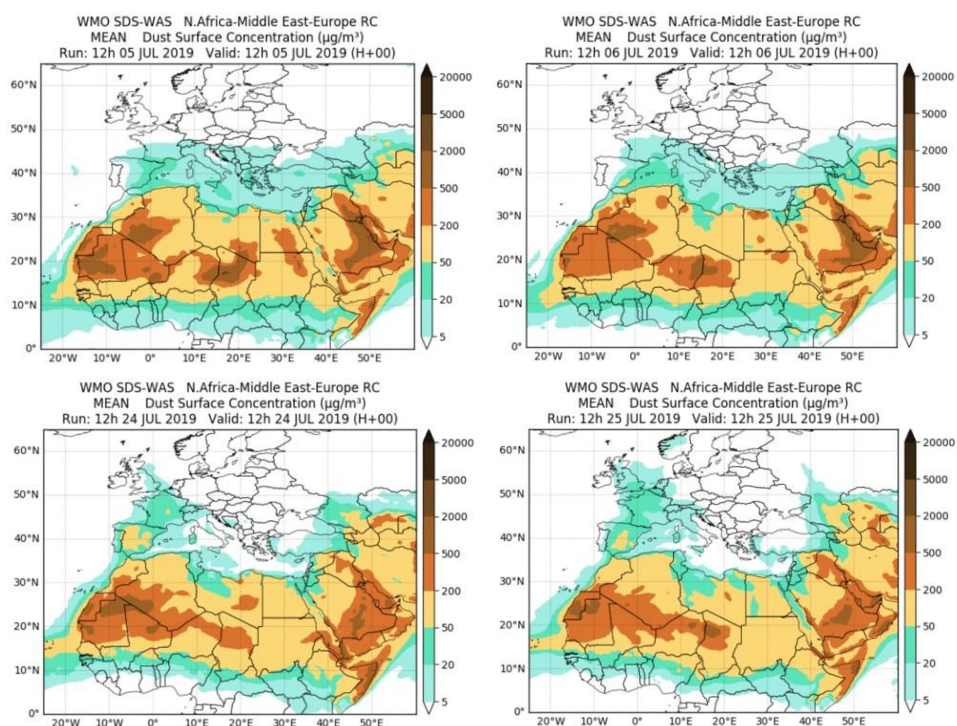
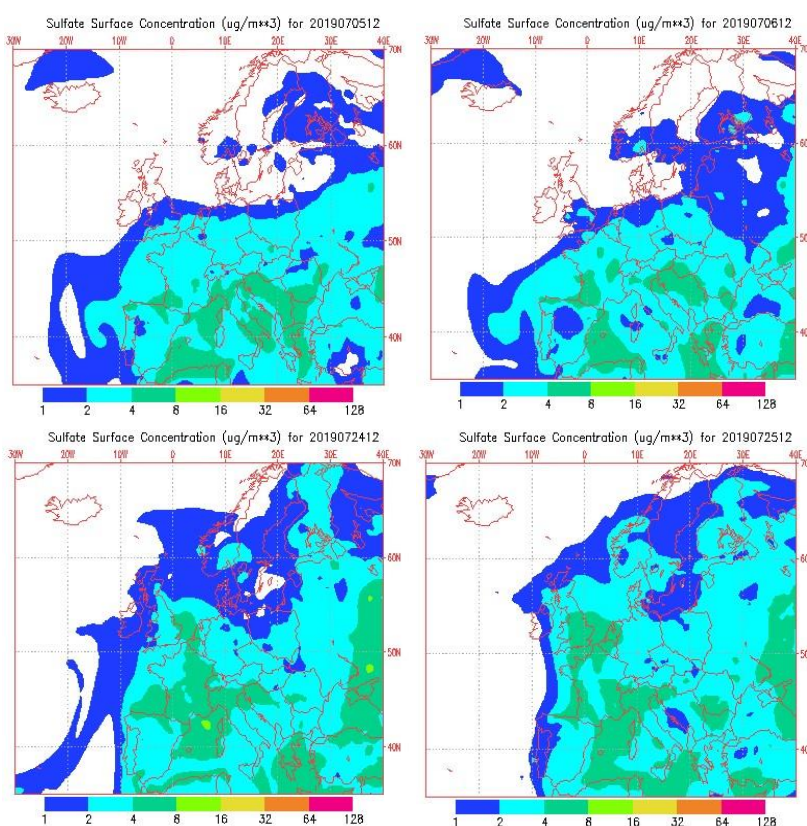


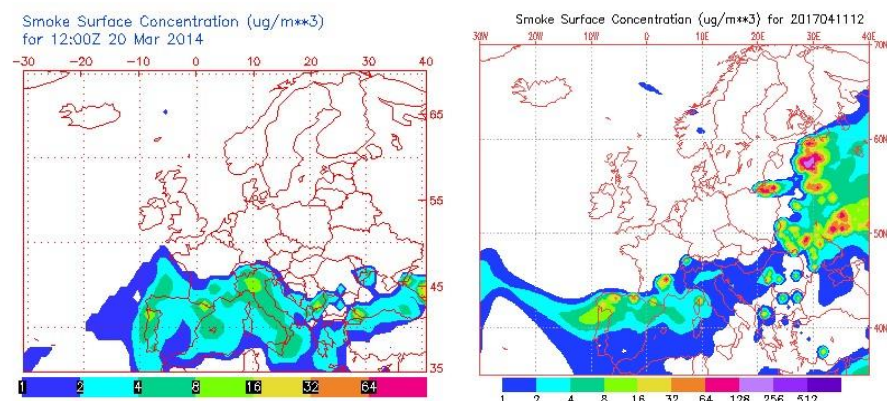
Figure S11. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s⁻¹) in BCN_UB.



S12. Mean dust surface concentration ($\mu\text{g m}^{-3}$) on four days in July 2019 with documented dust events in BCN_UB.



S13. Sulphate surface concentration ($\mu\text{g m}^{-3}$) on four days in July 2019.



S14. Smoke surface concentration ($\mu\text{g m}^{-3}$) on two days with documented forest fires in BCN_UB.

Table S7. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in BUD_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	SO ₂	NO	NO ₂	O ₃	CO
1	169 (16)	18	26	0	13	84	17
2	52	11	6	4	11	0	13
3	114	37	27	20	27	0	33
4	250 (114, 39)	7	11	0	0	2	7
5	20	8	15	29	28	3	17
UNDETERMINED		19	15	47	21	11	12

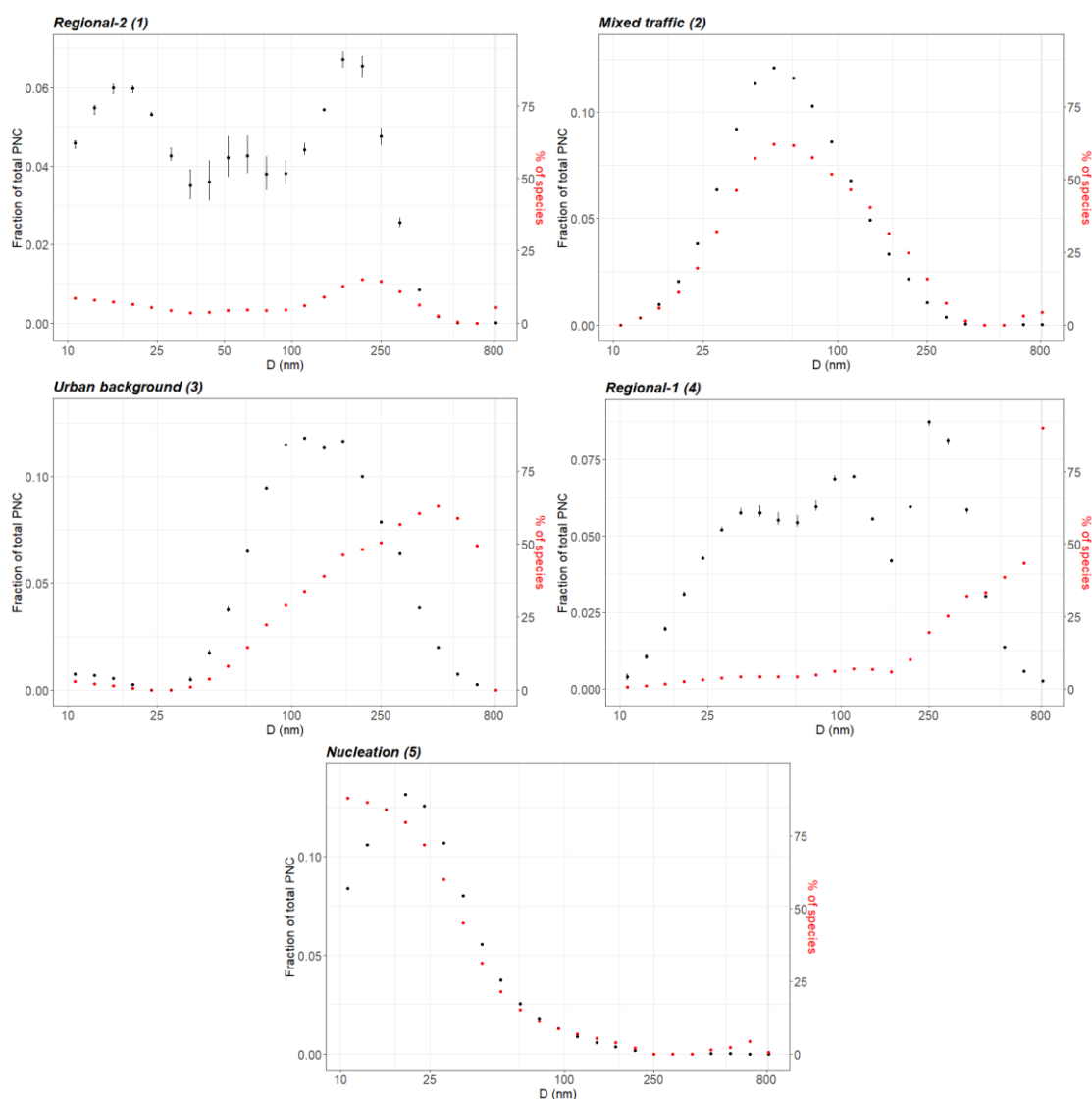


Figure S15. Particle number size distributions of each factor identified by PMF (before splitting) in BUD_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

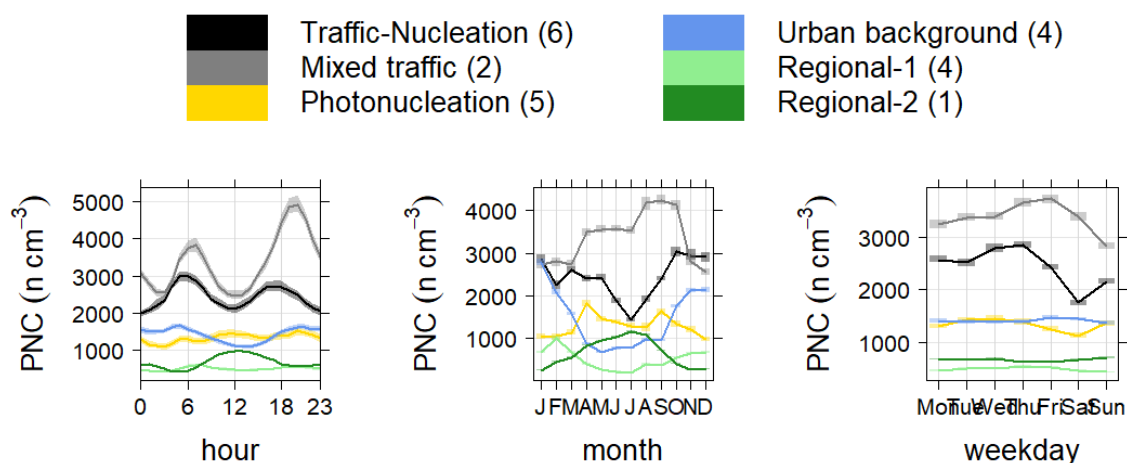


Figure S16. Daily, monthly and weekly patterns of the sources identified (after splitting) in BUD_UB.

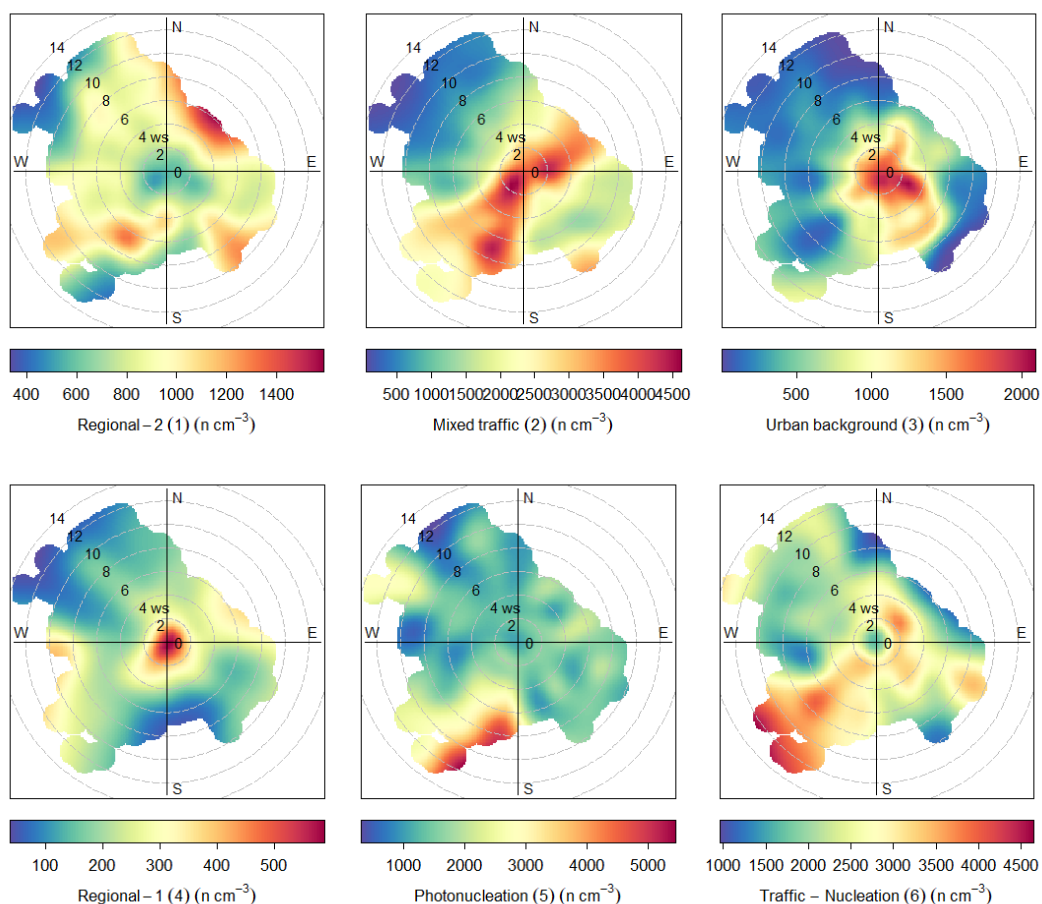


Figure S17. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in BUD_UB.

Table S8. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in DRE_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	SO ₂	NO	NO ₂	O ₃
1	232 (48)	15	15	0	6	32
2	28	6	10	9	17	0
3	119 (18)	31	19	11	23	0
4	11	1	8	2	1	7
5	54	4	0	3	17	0
6	291	28	8	8	14	0
UNDETERMINED		15	40	67	22	61

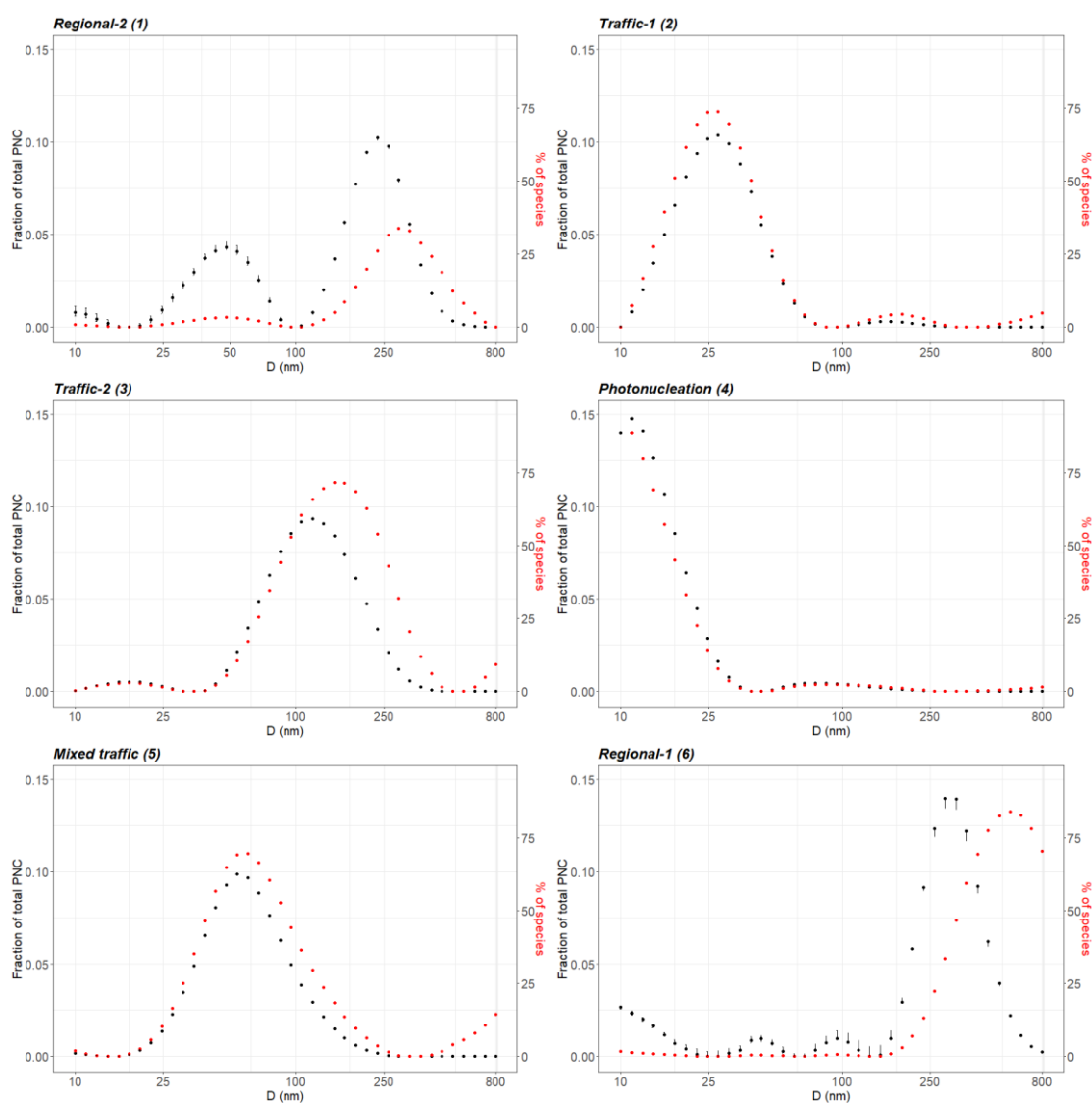


Figure S18. Particle number size distributions of each factor identified by PMF (before splitting) in DRE_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

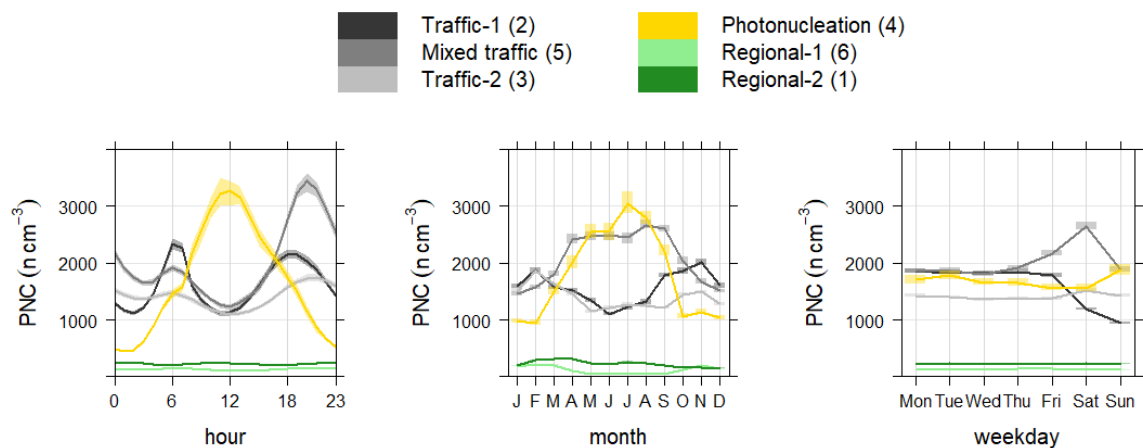


Figure S19. Daily, monthly and weekly patterns of the sources identified (after splitting) in DRE_UB.

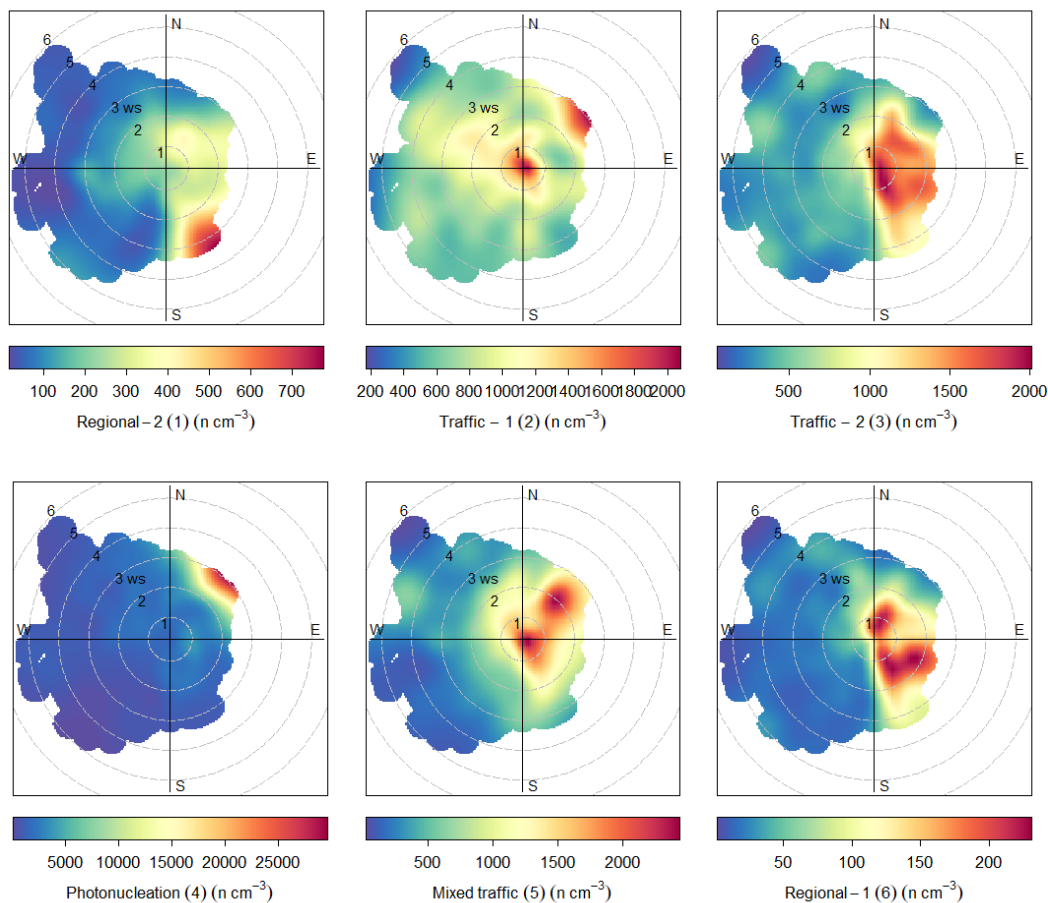


Figure S20. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s⁻¹) in DRE_UB.

Table S9. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in GRA_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	SO ₂	NO	NO ₂	O ₃	CO
1	43	17	3	5	0	12	0	9
2	98	37	14	0	15	37	0	32
3	23	8	0	3	3	9	0	16
4	13 (57)	0	6	9	1	5	28	3
5	175 (27)	19	37	10	1	9	2	14
UNDETERMINED		18	40	74	81	28	70	26

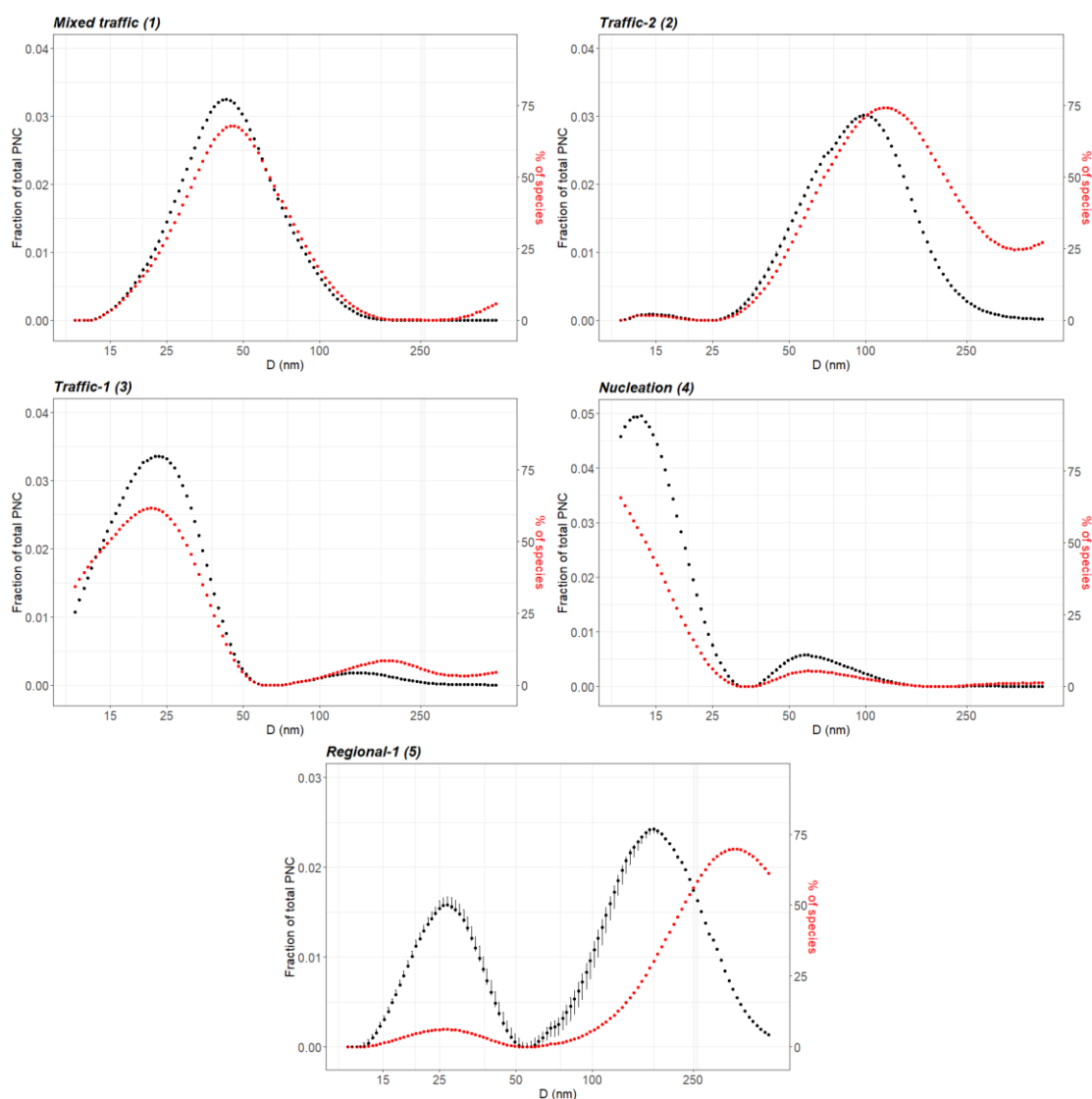


Figure S21. Particle number size distributions of each factor identified by PMF (before splitting) in GRA_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

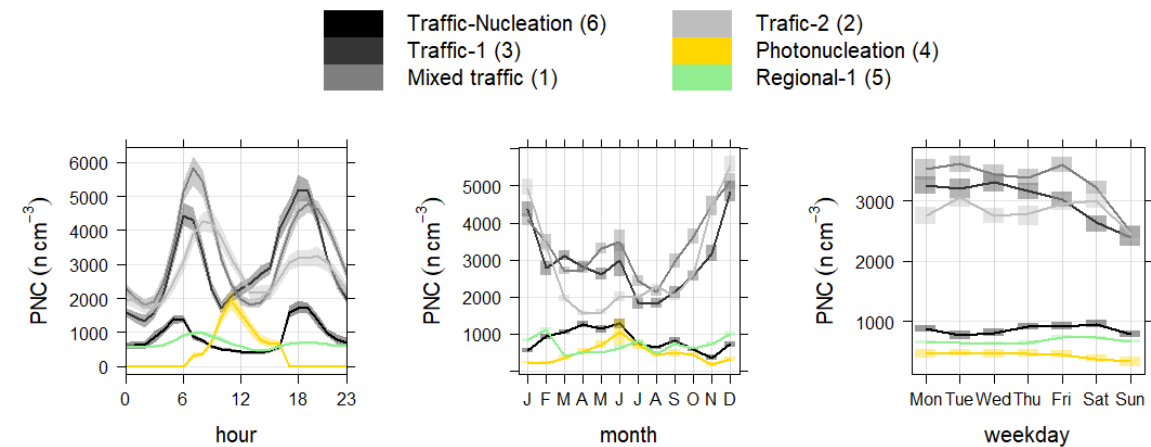


Figure S22. Daily, monthly and weekly patterns of the sources identified (after splitting) in GRA_UB.

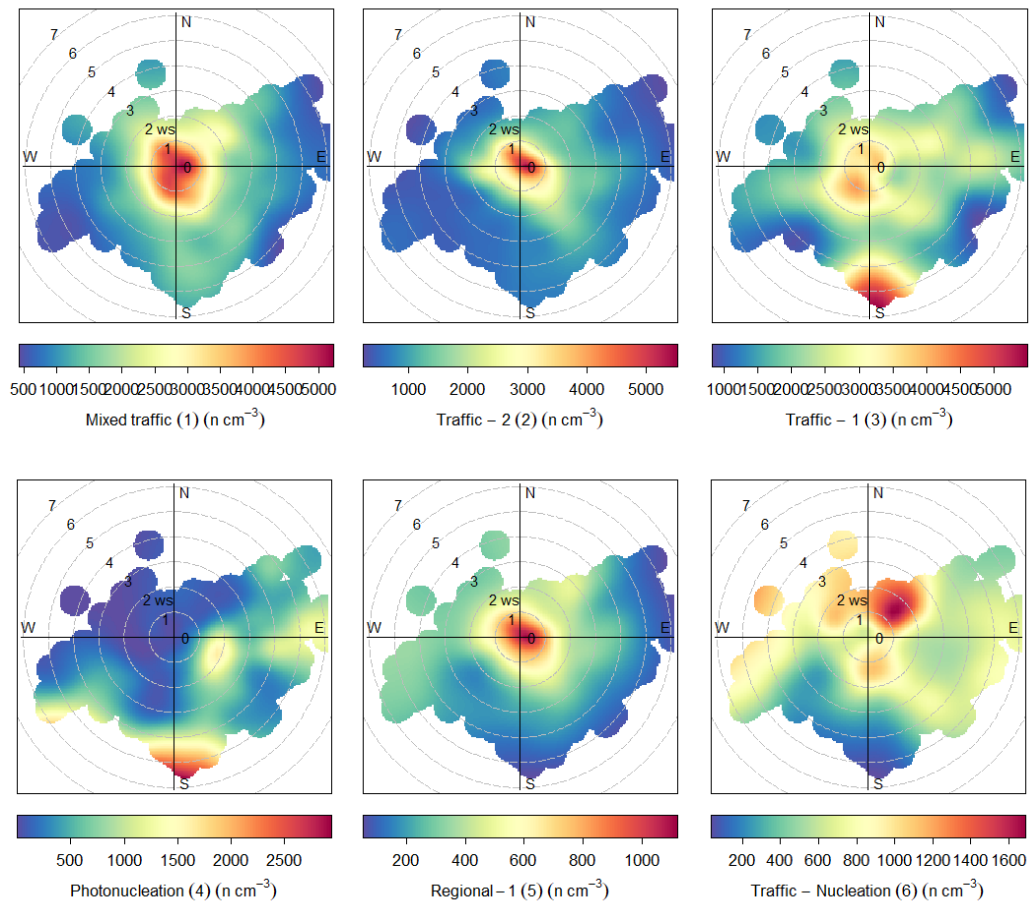


Figure S23. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in GRA_UB.

Table S10. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in HEL_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	SO ₂	NO ₂	O ₃	CO
1	45	11	25	0	15
2	80	6	5	57	20
3	16	6	15	2	23
4	200 (28)	7	8	0	22
UNDETERMINED		70	47	41	20

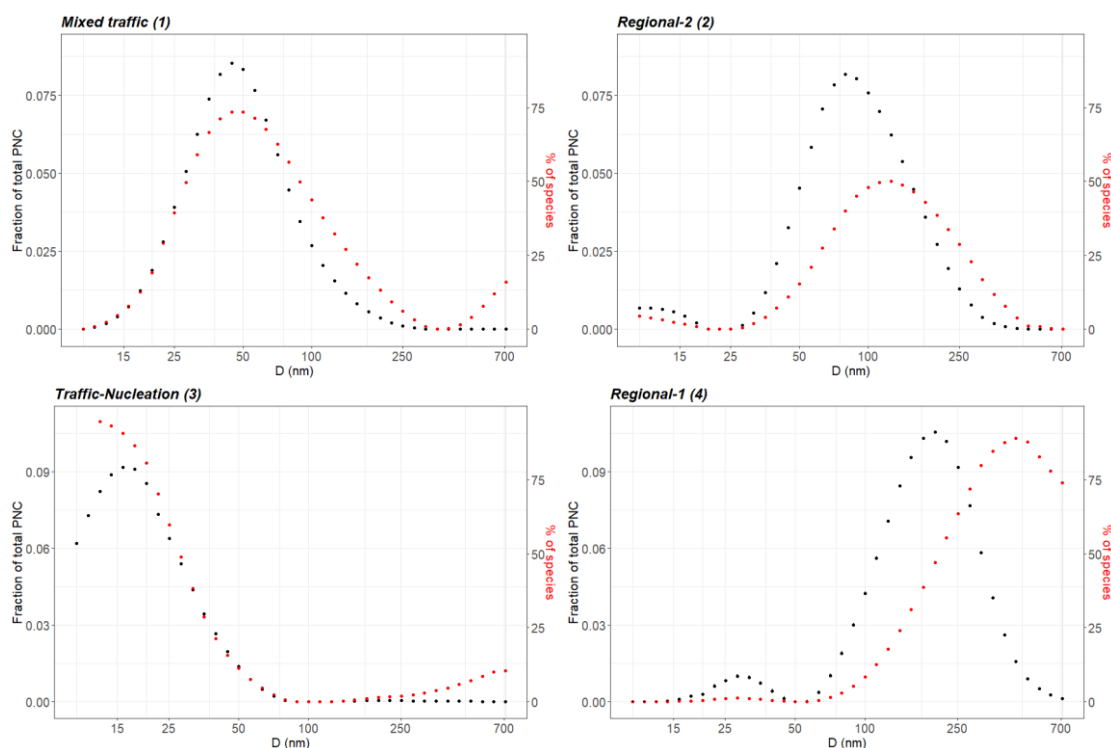


Figure S24. Particle number size distributions of each factor identified by PMF in HEL_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

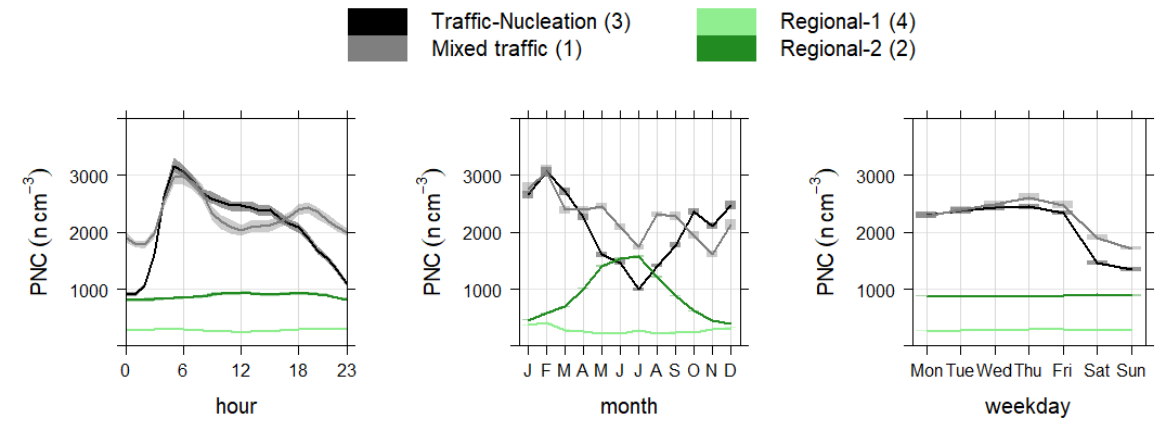


Figure S25. Daily, monthly and weekly patterns of the sources identified in HEL_UB.

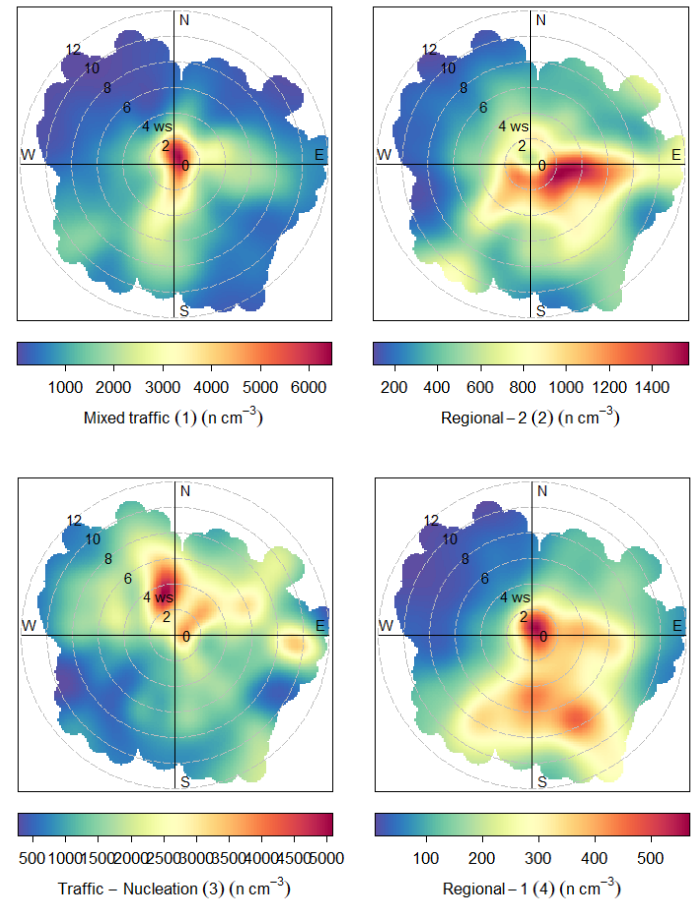


Figure S26. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in HEL_UB.

Table S11. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in LEC_UB. UNDETERMINED refers to relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	SO ₂	NO	NO ₂	O ₃
1	260	17	1	0	9	0
2	60	24	0	0	31	3
3	27	4	0	0	9	0
4	106	37	10	0	16	0
5	208 & 48 (10)	0	35	1	0	84
6	11	1	3	1	2	1
UNDETERMINED		17	51	97	33	12

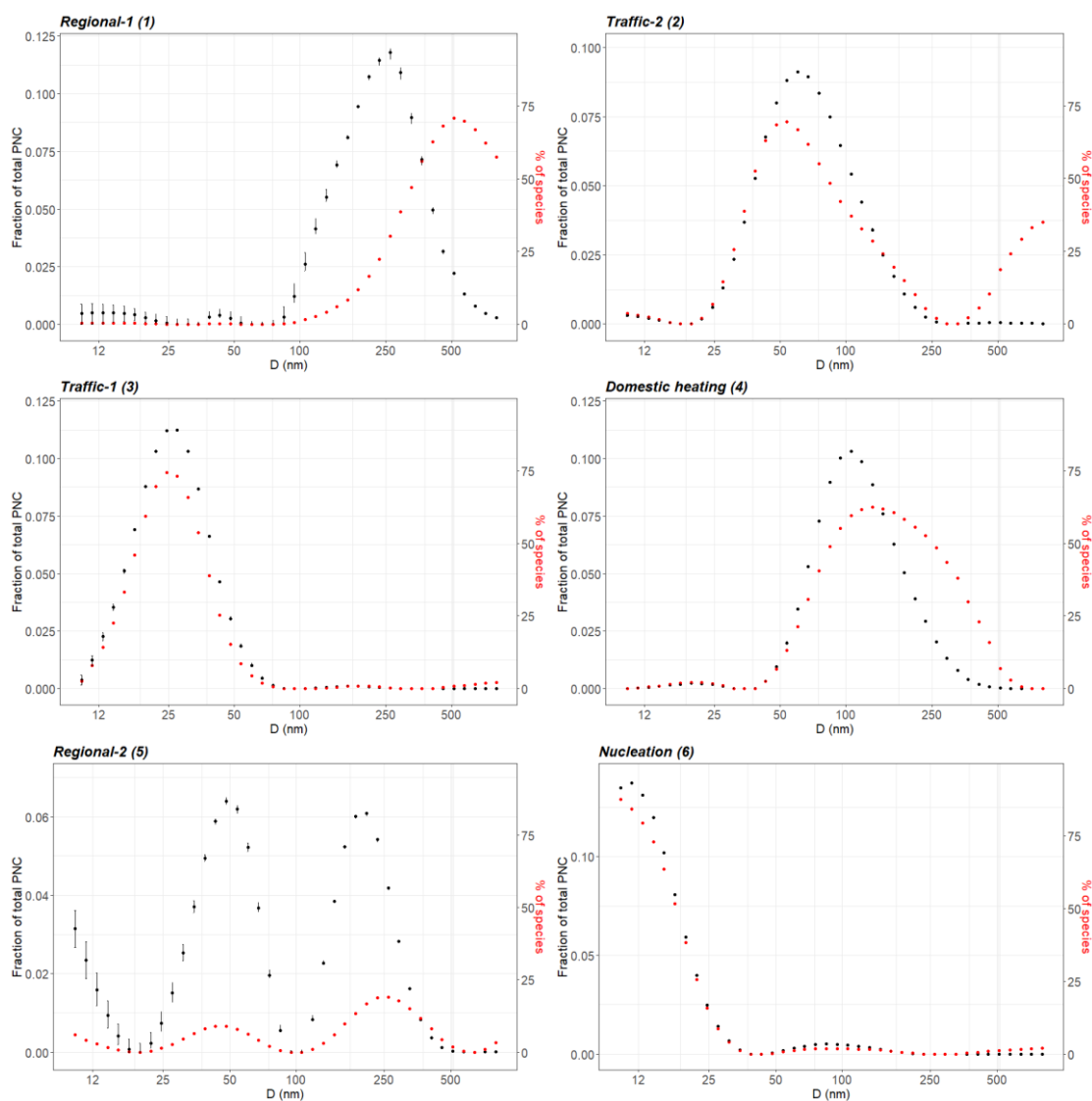


Figure S27. Particle number size distributions of each factor identified by PMF (before splitting) in LEC_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

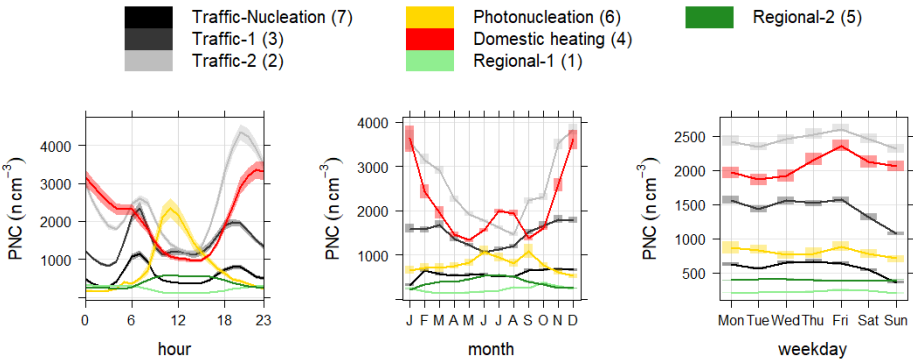


Figure S28. Daily, monthly and weekly patterns of the sources identified (after splitting) in LEC_UB.

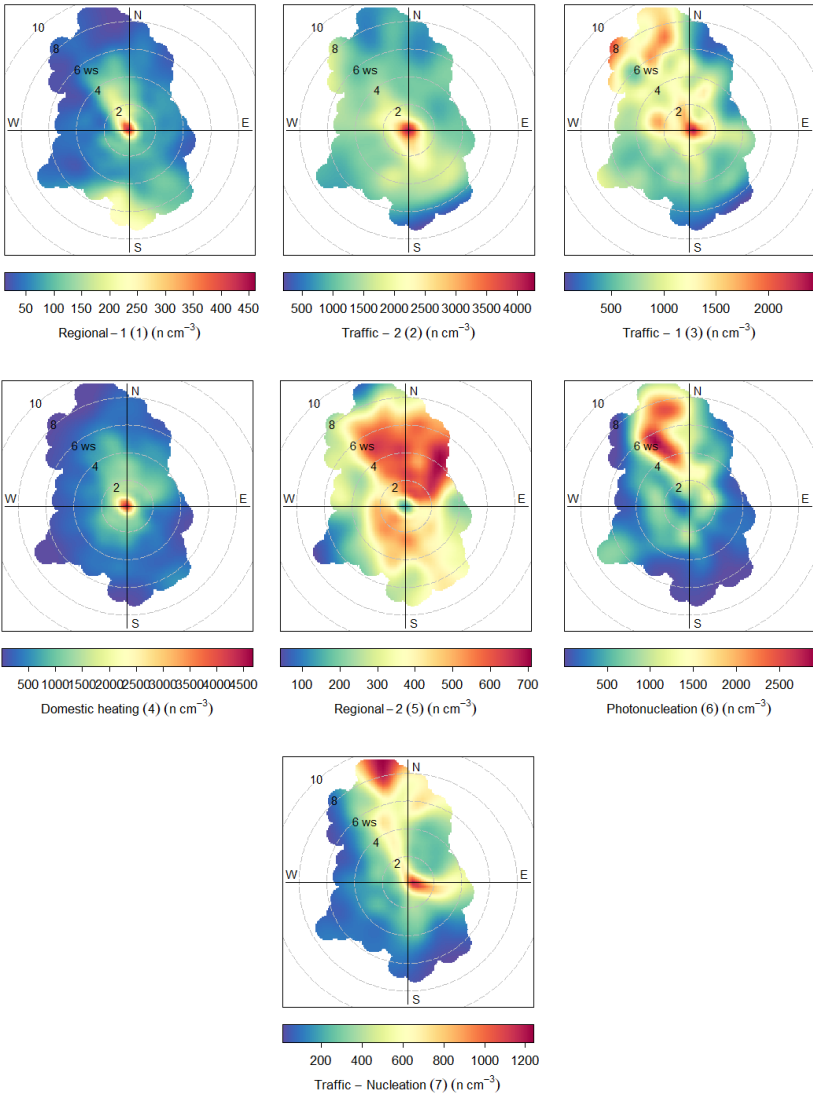


Figure S29. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in LEC_UB.

Table S12. Size mode of each factor identified by PMF (before applying splitting methodology) in LEI_UB. This site does not have ancillary pollutants data.

Factor	Mode (nm)
1	13
2	54
3	232
4	364 (95, 14)
5	106 (20)
6	28

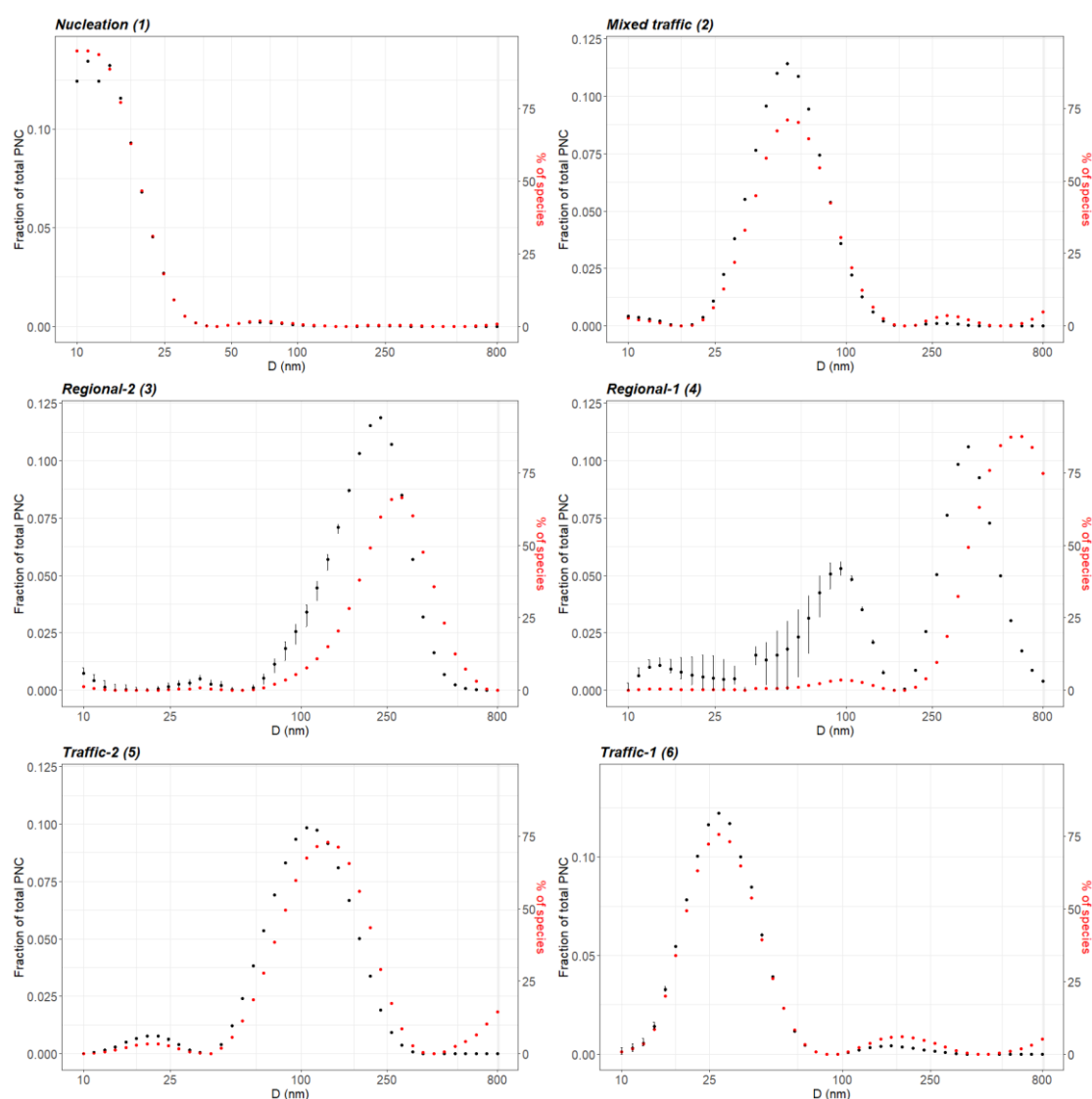


Figure S30. Particle number size distributions of each factor identified by PMF (before splitting) in LEI_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

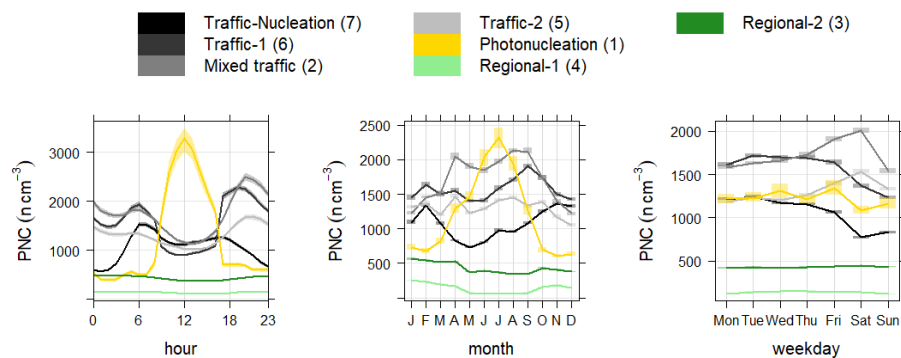


Figure S31. Daily, monthly and weekly patterns of the sources identified (after splitting) in LEI_UB.

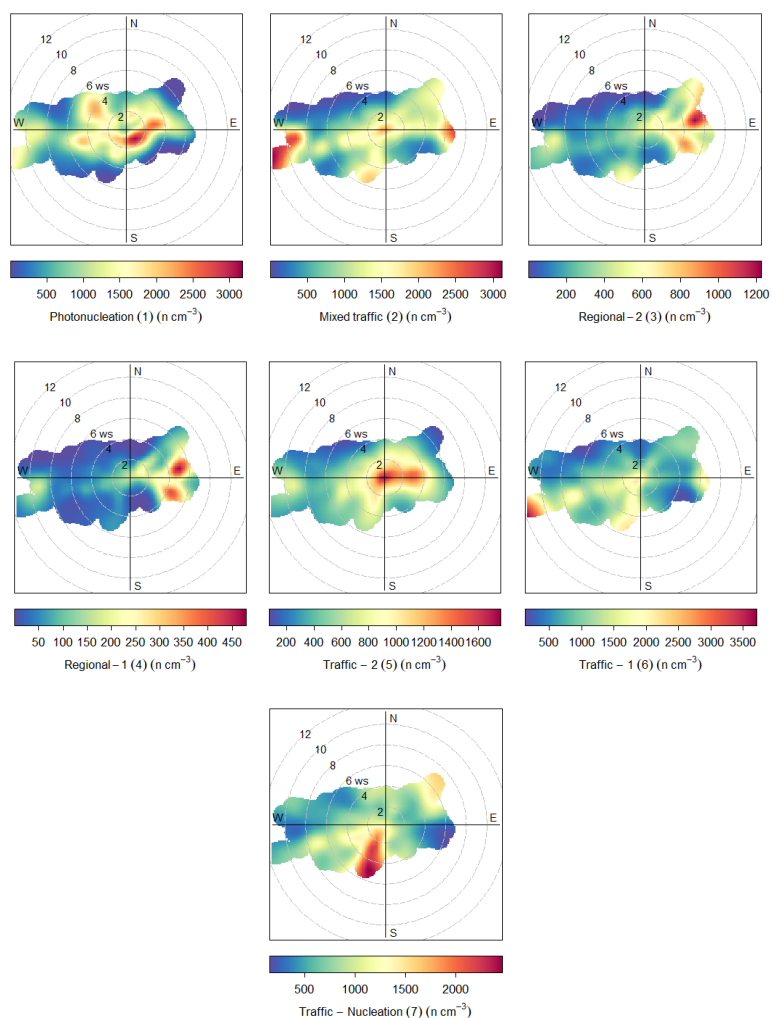


Figure S32. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in LEI_UB. No meteorological data is measured in this site, thus wind speed and wind direction from LEI_TR were used for the polar plots.

Table S13. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in LND_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM _{2.5}	SO ₂	NO	NO ₂	O ₃	CO
1	87	43	34	27	1	28	0	35
2	37	22	6	19	4	42	0	26
3	255 (65)	14	38	10	1	14	0	16
4	22	1	3	10	1	1	32	7
UNDETERMINED		21	19	35	92	15	68	16

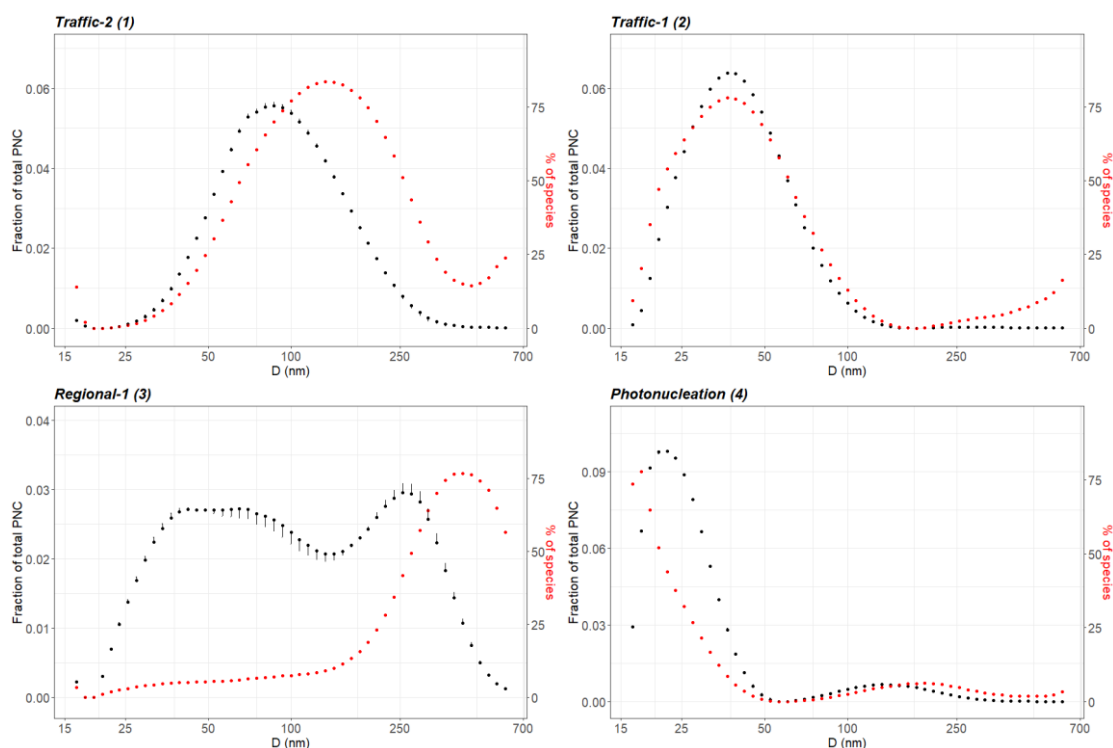


Figure S33. Particle number size distributions of each factor identified by PMF in LND_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

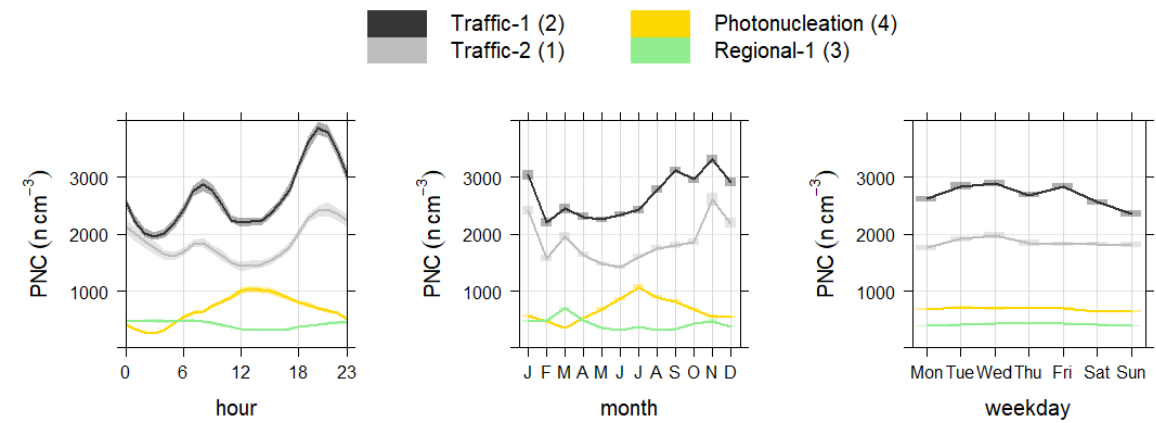


Figure S34. Daily, monthly and weekly patterns of the sources identified in LND_UB.

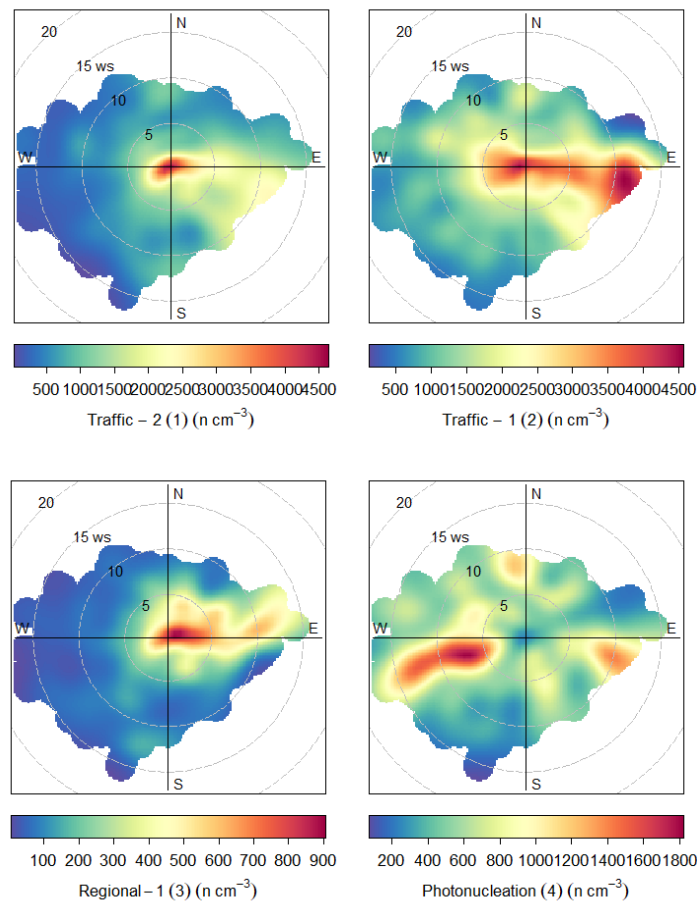


Figure S35. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in LND_UB.

Table S14. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in MAD_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	SO ₂	NO	NO ₂	O ₃	CO
1	69	18	16	6	1	19	0	3
2	37	7	0	21	7	16	0	18
3	151 (20)	5	32	31	0	3	51	23
4	188 (24)	38	23	13	21	27	0	26
5	19	0	0	0	0	3	3	0
UNDETERMINED		32	29	29	71	33	47	29

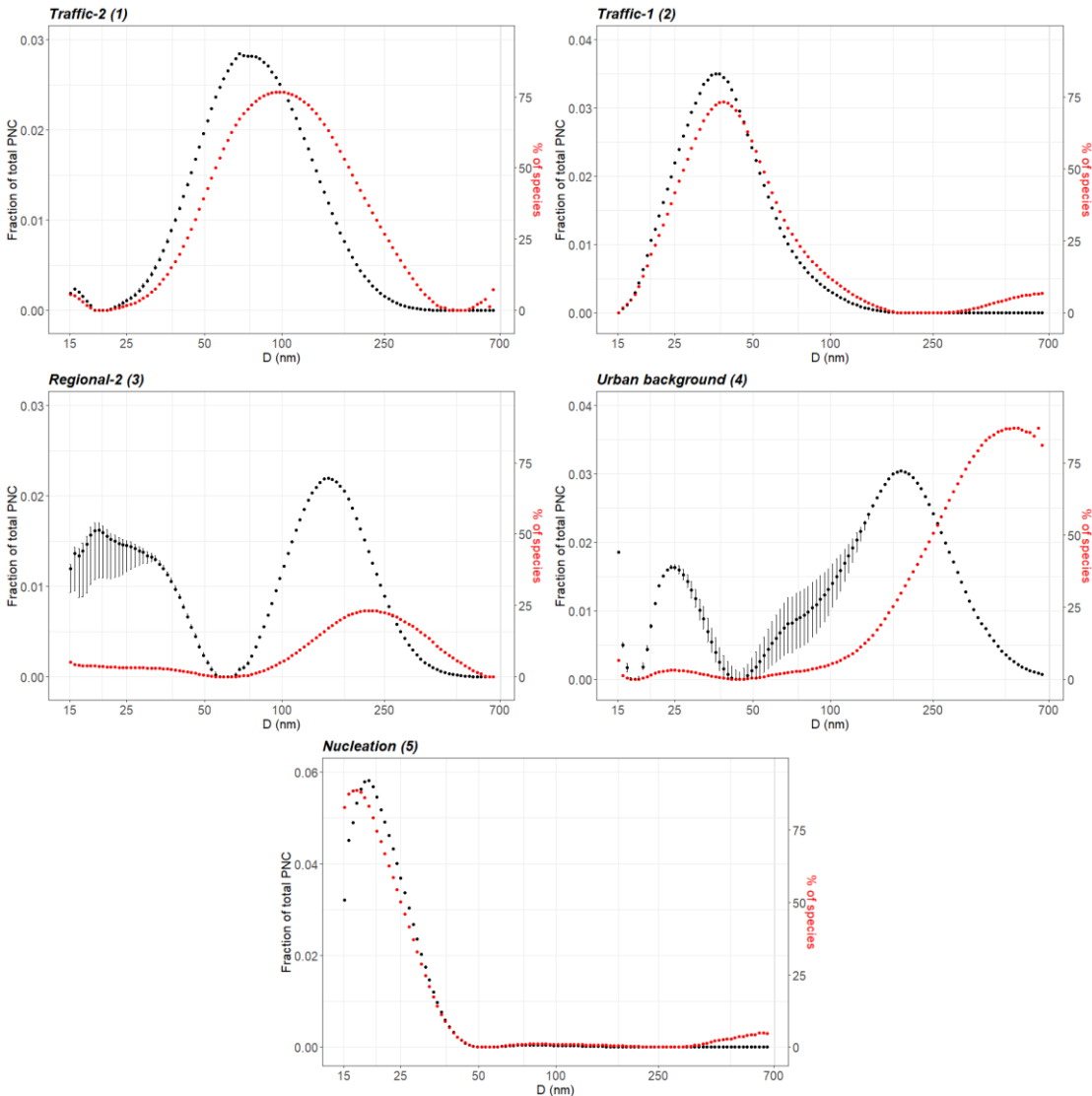


Figure S36. Particle number size distributions of each factor identified by PMF (before splitting) in MAD_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

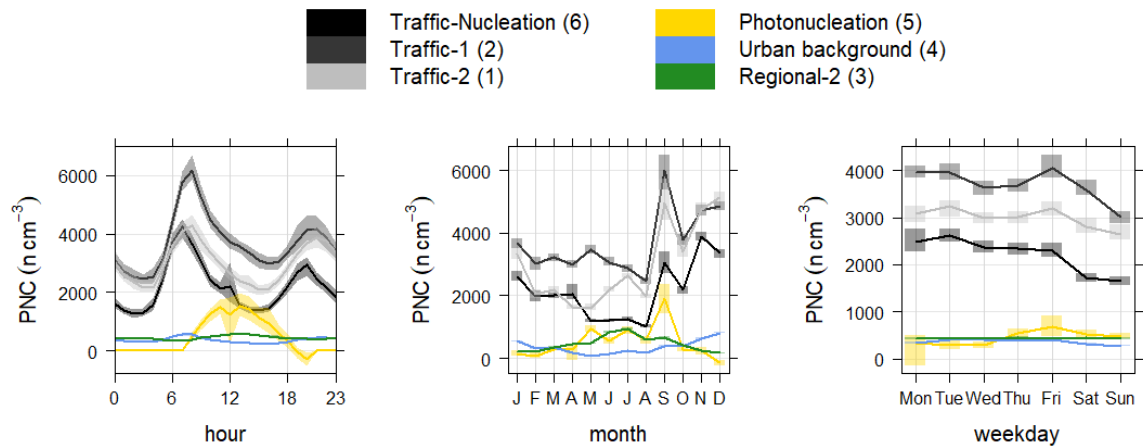


Figure S37. Daily, monthly and weekly patterns of the sources identified (after splitting) in MAD_UB.

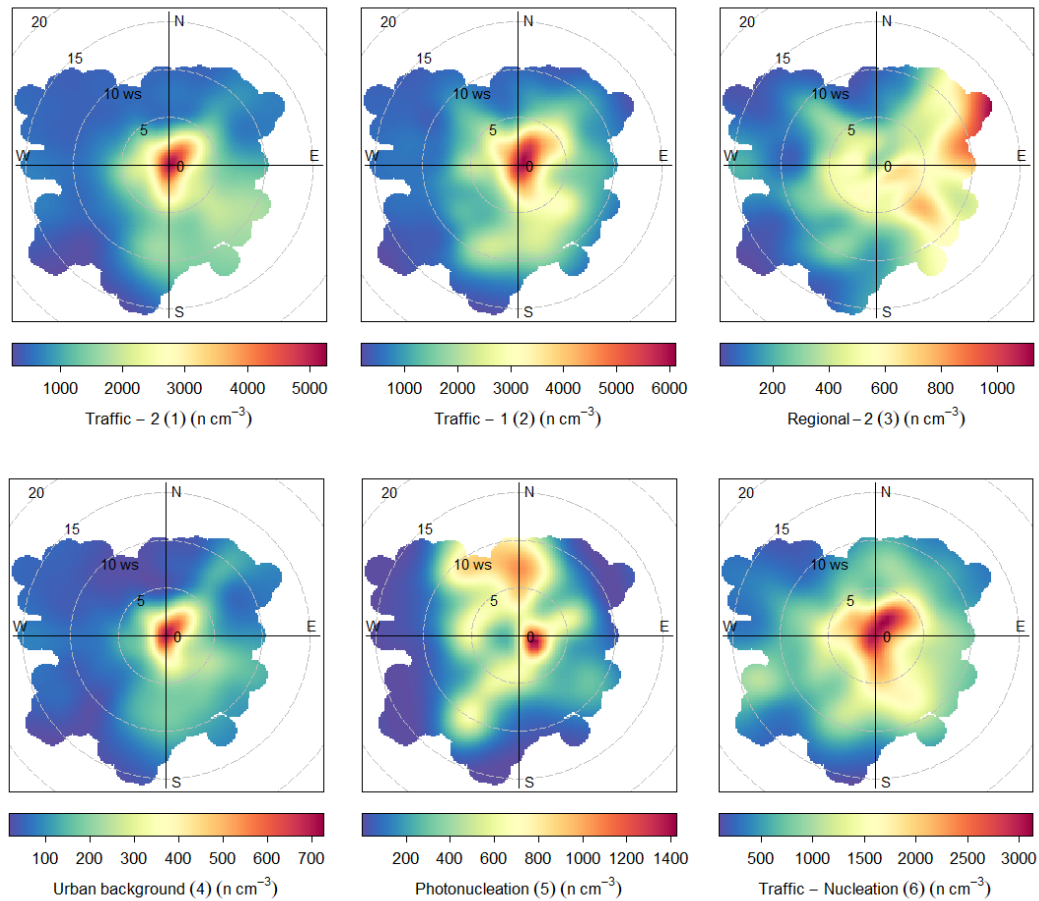


Figure S38. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s⁻¹) in MAD_UB.

Table S15. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in MUL_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	NO	NO ₂
1	50	0	0	16
2	15	0	0	3
3	29	13	0	13
4	322 (78, 19)	47	2	18
5	188 (36)	0	0	2
6	88 (20)	20	0	22
UNDETERMINED		20	97	26

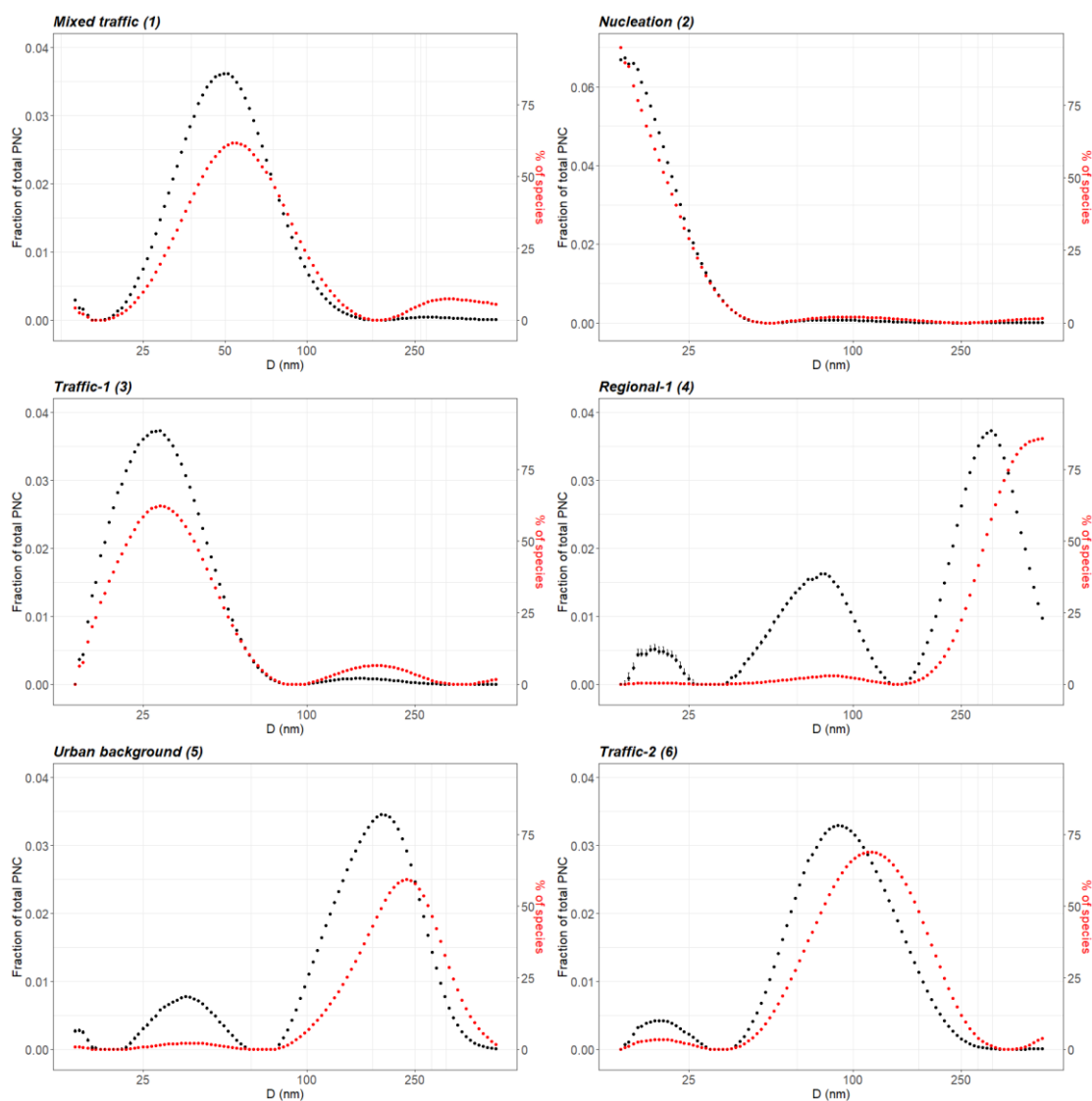


Figure S39. Particle number size distributions of each factor identified by PMF (before splitting) in MUL_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

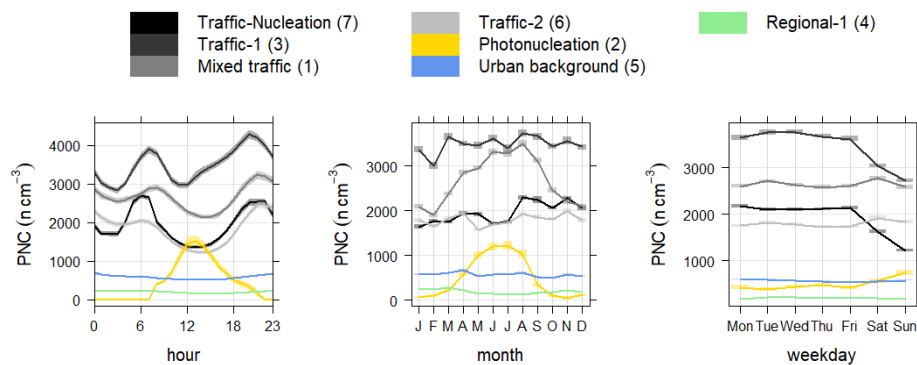


Figure S40. Daily, monthly and weekly patterns of the sources identified (after splitting) in MUL_UB.

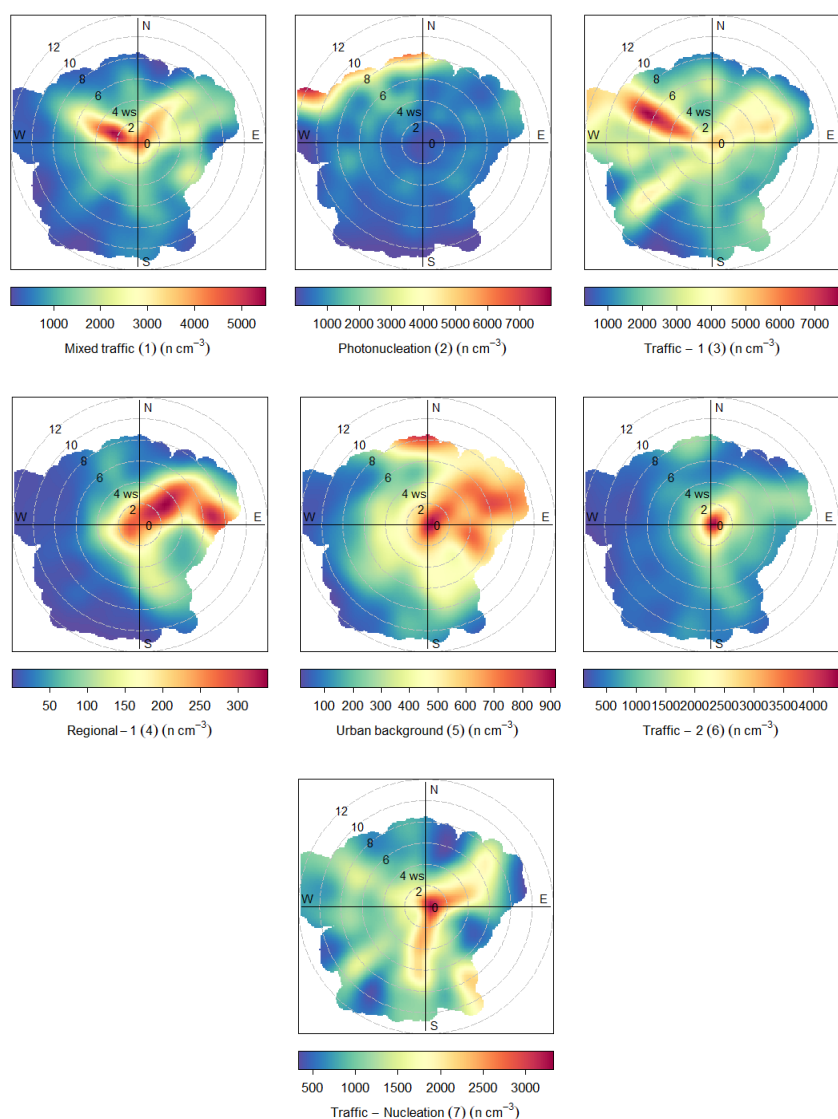


Figure S41. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in MUL_UB.

Table S16. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in ROC_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM _{2.5}	SO ₂	NO	NO ₂	O ₃
1	249	17	43	0	0	9	50
2	67	27	0	0	0	1	0
3	35	1	0	0	0	1	11
4	246 (20)	13	2	3	1	0	0
5	41	19	27	30	31	54	0
6	18	6	0	0	1	2	0
UNDETERMINED		17	28	67	67	33	39

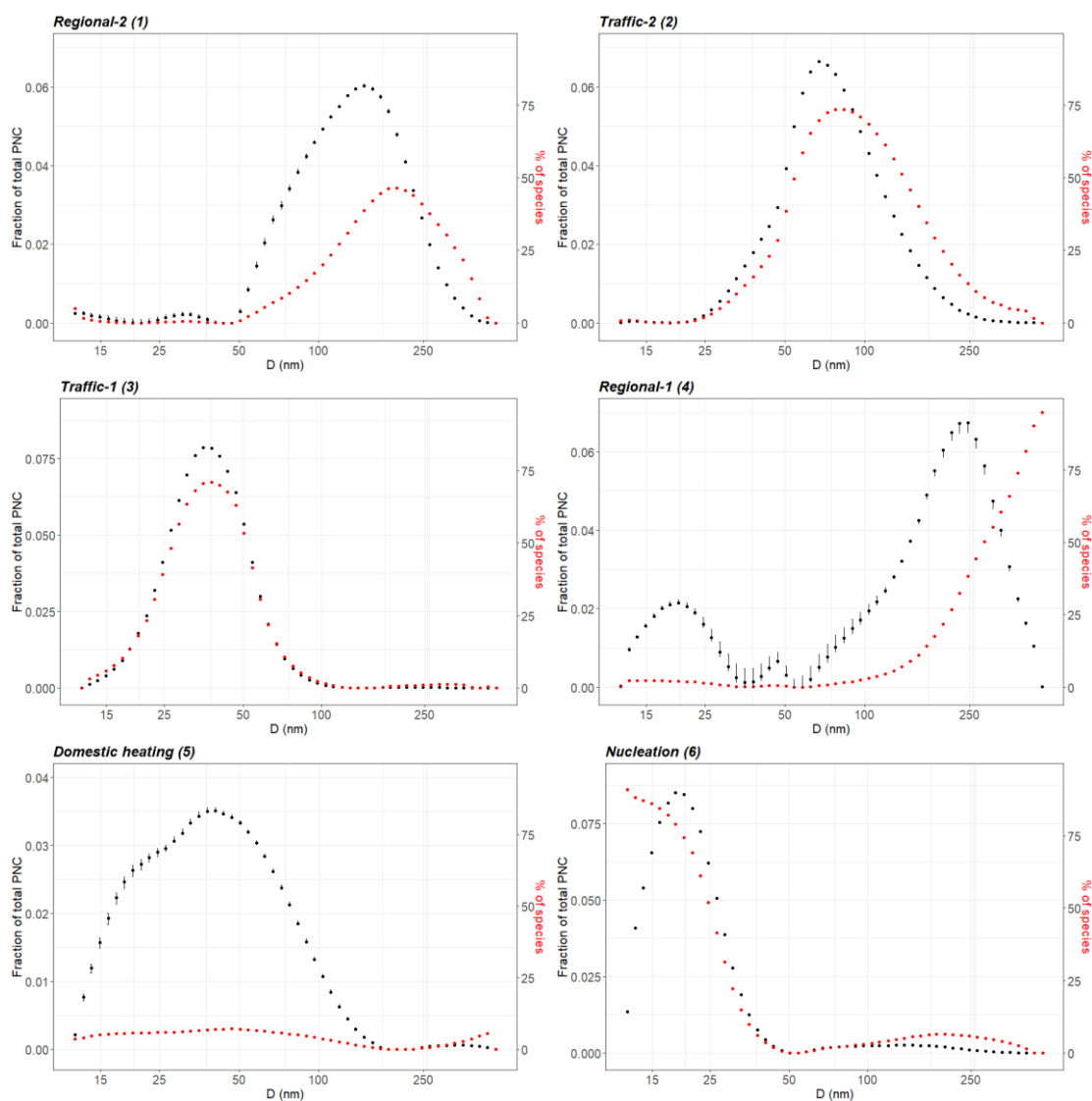


Figure S42. Particle number size distributions of each factor identified by PMF (before splitting) in ROC_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

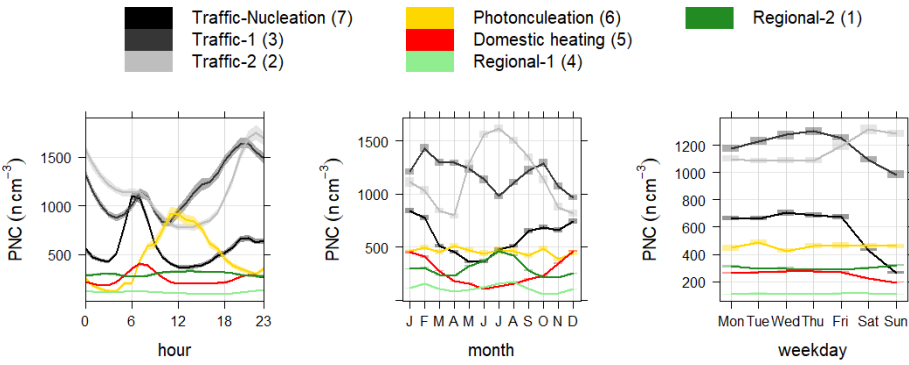


Figure S43. Daily, monthly and weekly patterns of the sources identified (after splitting) in ROC_UB.

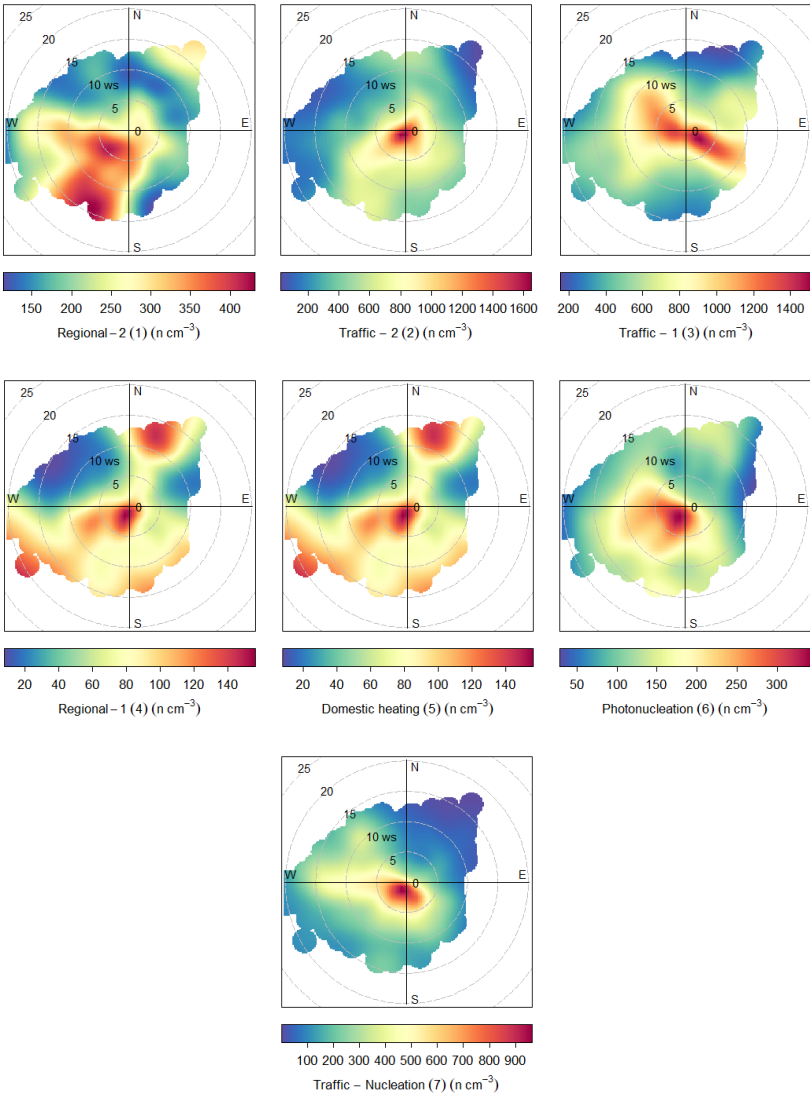


Figure S44. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in ROC_UB.

Table S17. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in ZUR_UB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	SO ₂	NO	NO ₂	O ₃	CO
1	20	0	0	0	0	0	0	0
2	188 (32)	5	16	0	0	4	57	14
3	31	10	4	18	5	21	0	16
4	55	13	6	6	0	17	0	12
5	106	36	25	17	2	22	0	26
6	300 (31)	25	35	19	3	20	0	18
UNDETERMINED		11	15	40	89	16	43	13

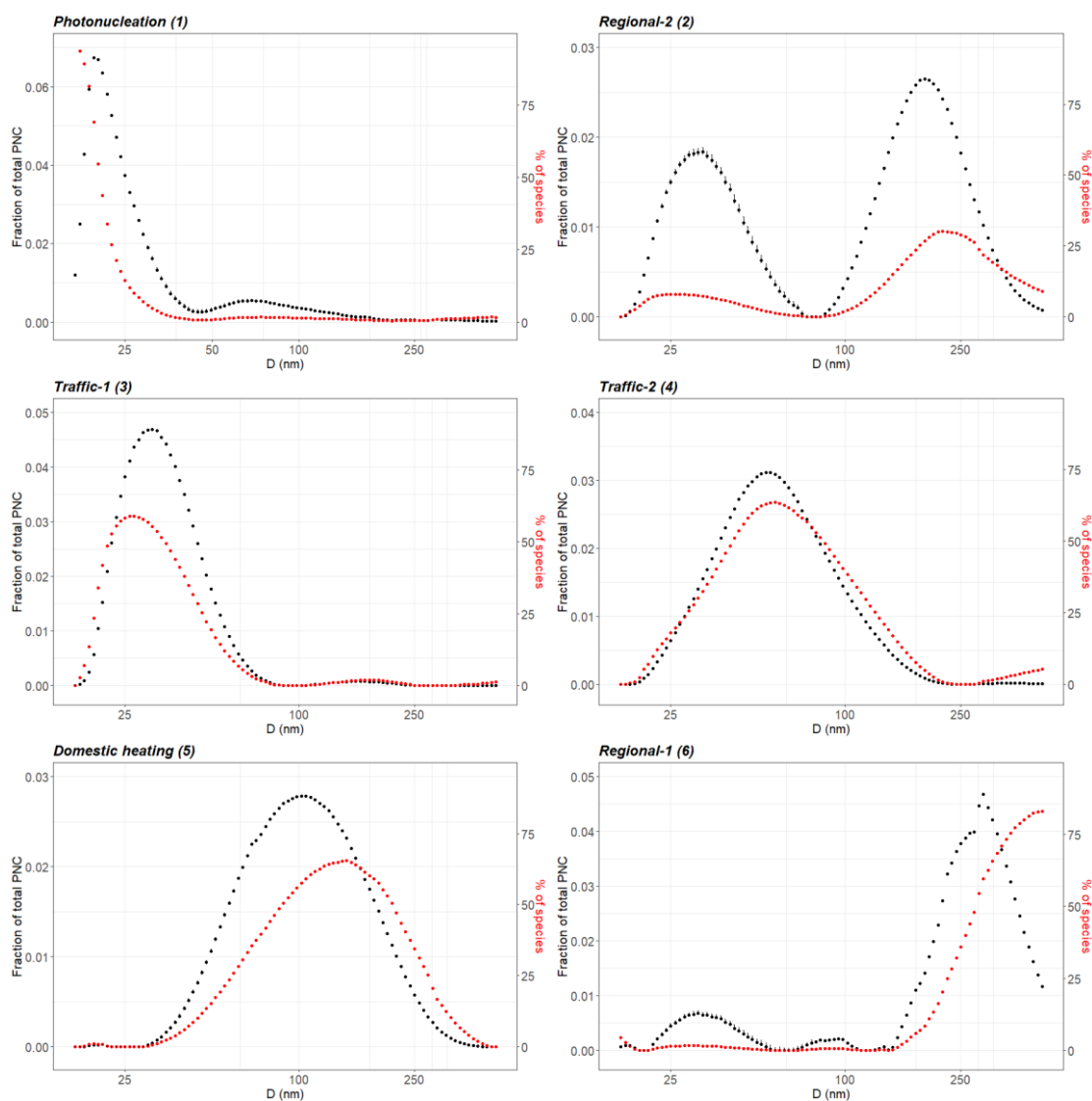


Figure S45. Particle number size distributions of each factor identified by PMF in ZUR_UB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

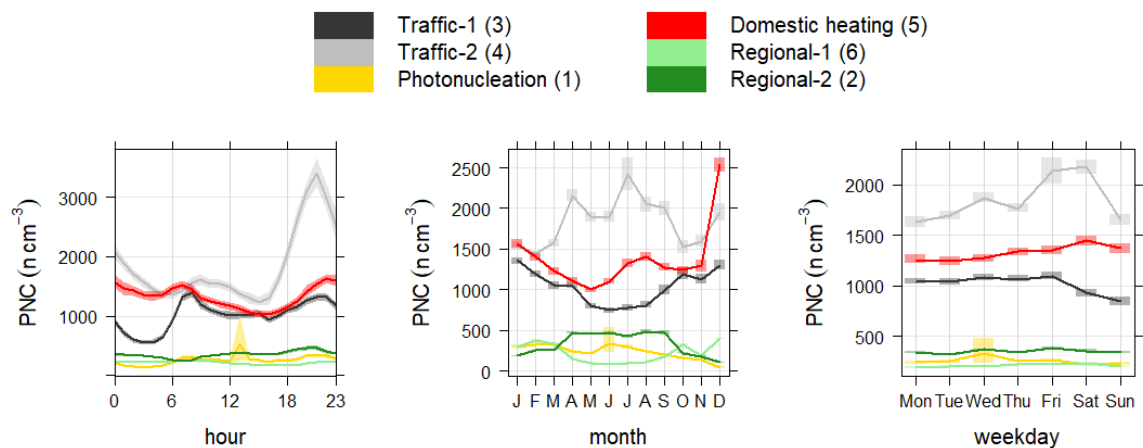


Figure S46. Daily, monthly and weekly patterns of the sources identified in ZUR_UB.

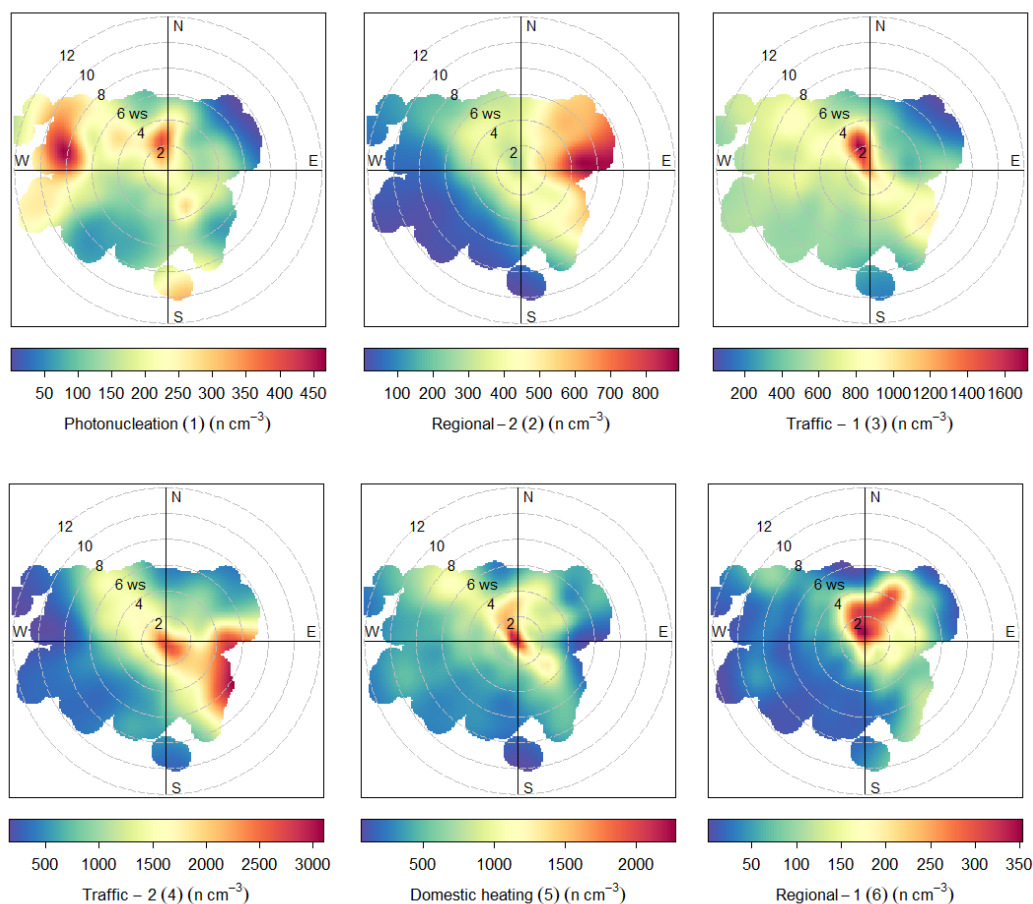


Figure S47. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in ZUR_UB.

Table S18. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in DRE_TR. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	SO ₂	NO	NO ₂	O ₃
1	85	27	25	6	26	0
2	16	14	4	43	4	0
3	38	17	9	3	9	4
4	28 & 208	3	17	0	18	54
5	260 (28)	25	31	4	30	0
UNDETERMINED		15	13	44	13	41

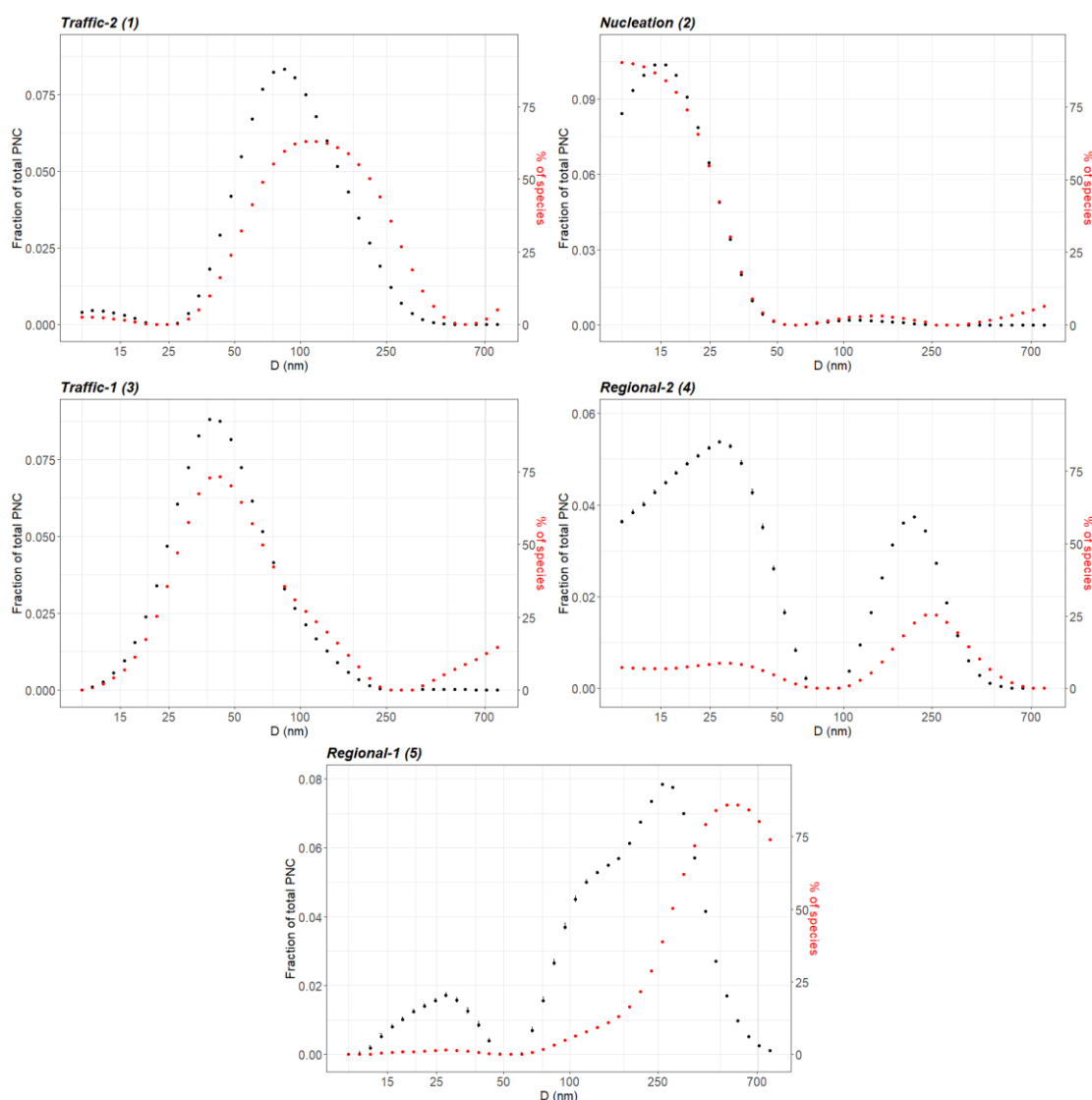


Figure S48. Particle number size distributions of each factor identified by PMF (before splitting) in DRE_TR. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

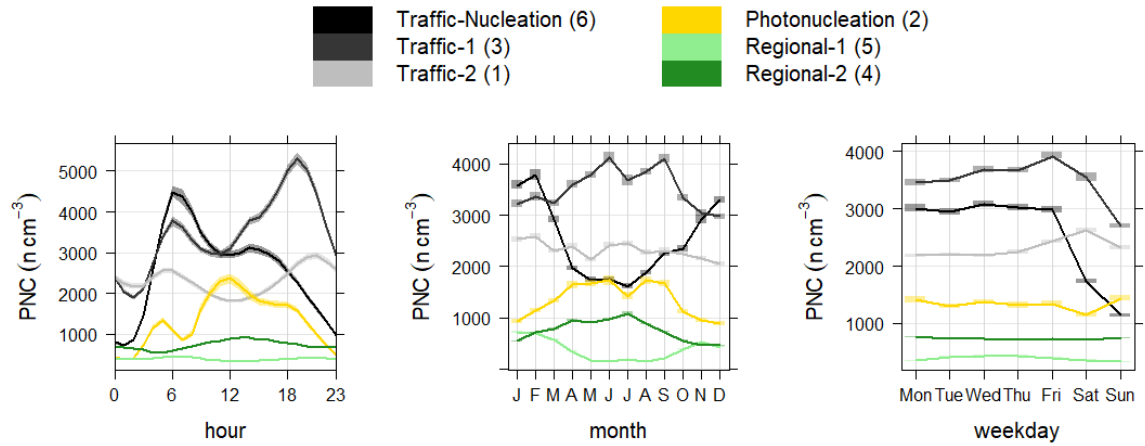


Figure S49. Daily, monthly and weekly patterns of the sources identified (after splitting) in DRE_TR.

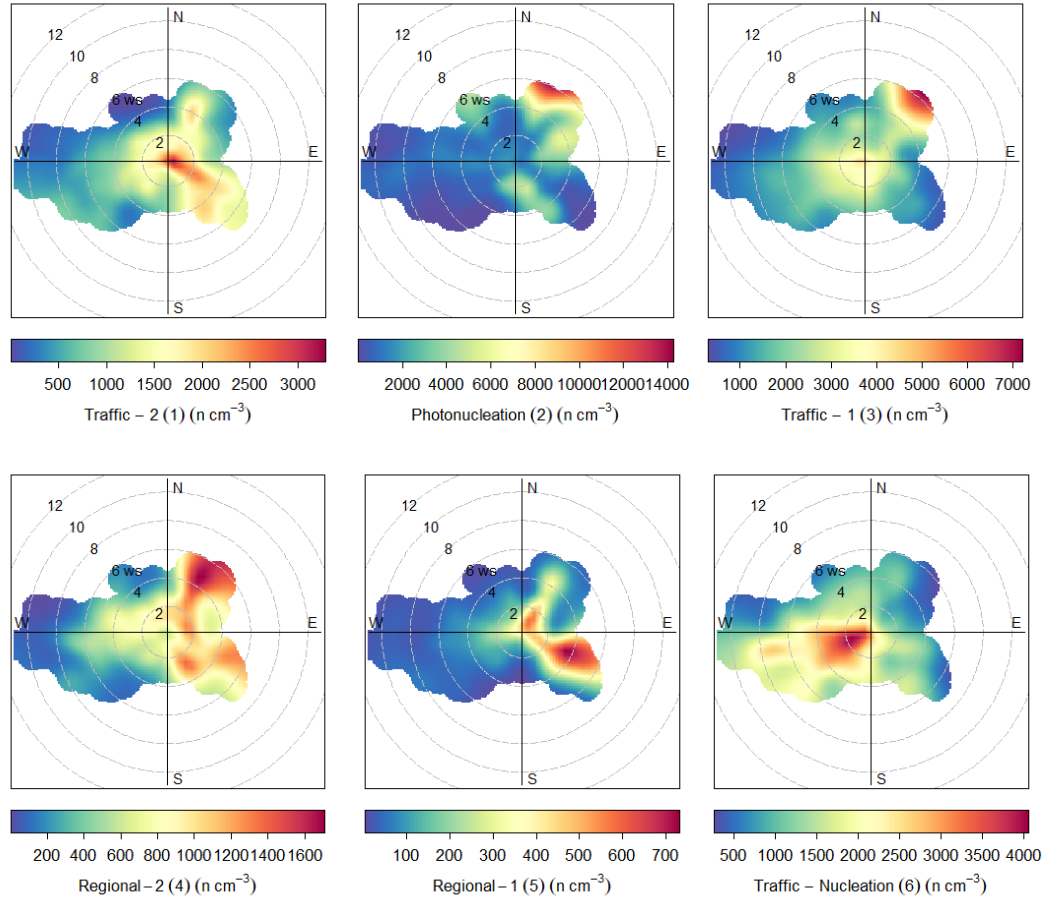


Figure S50. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in DRE_TR.

Table S19. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in HEL_TR. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃
1	30 (176)	3	29	26	20	0	13	89
2	213 (11)	13	6	30	7	0	4	0
3	13	5	0	0	2	8	11	2
4	55	16	1	0	3	2	13	0
UNDETERMINED		63	64	44	68	90	59	9

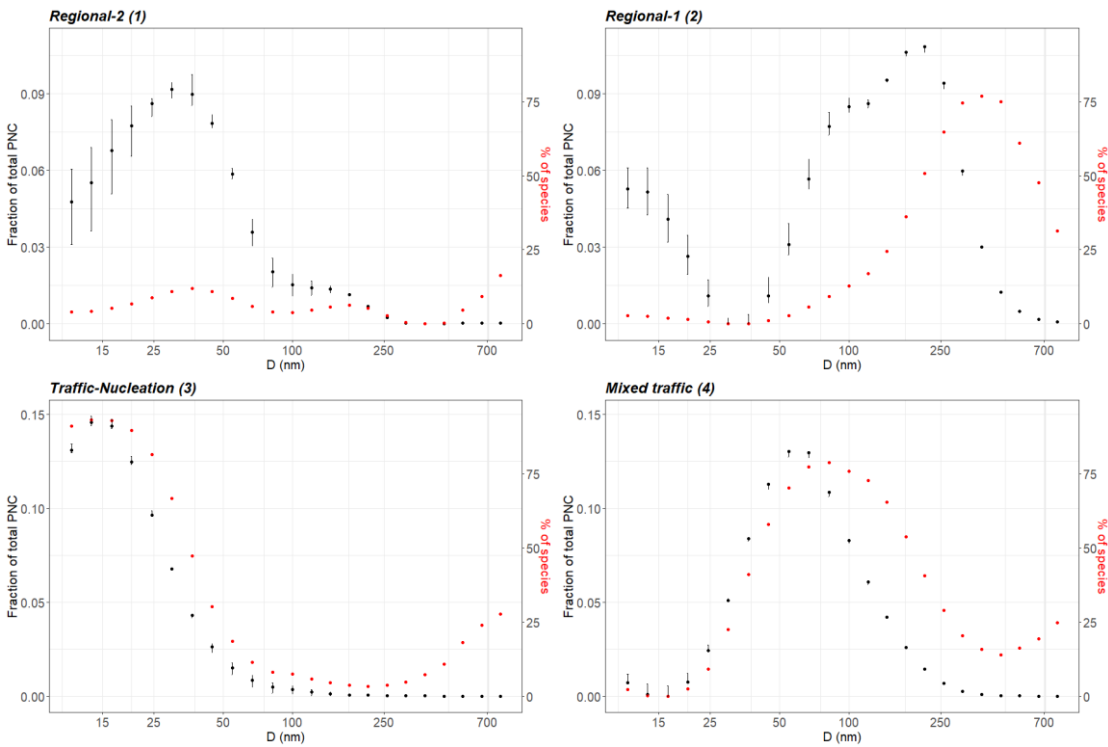


Figure S51. Particle number size distributions of each factor identified by PMF in HEL_TR. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

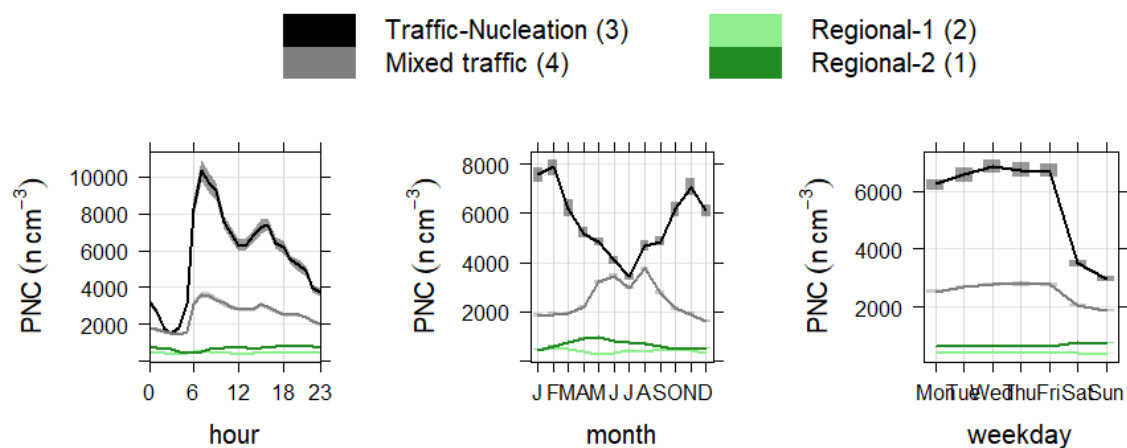


Figure S52. Daily, monthly and weekly patterns of the sources identified in HEL_TR.

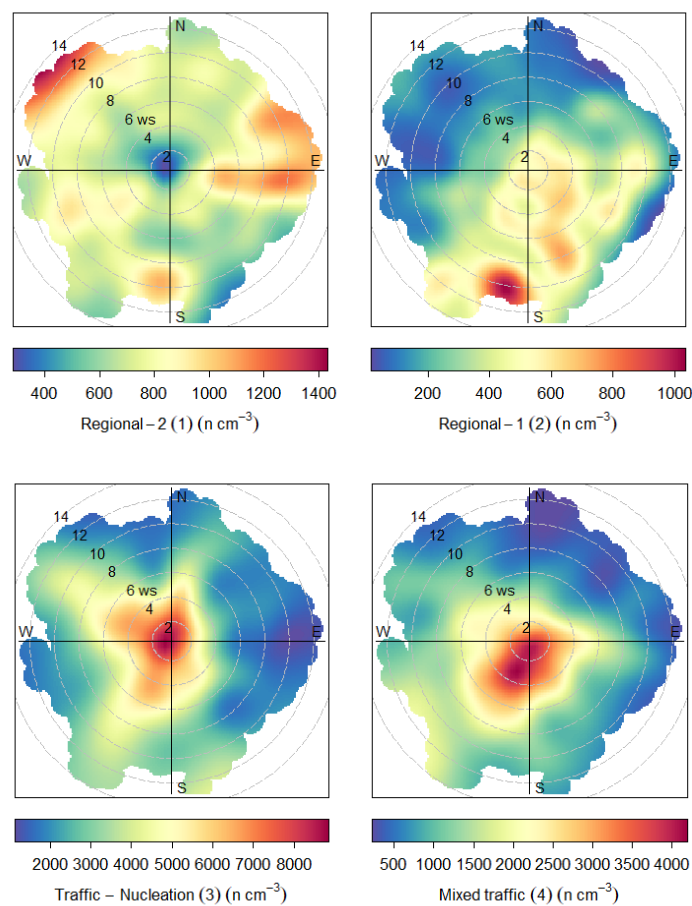


Figure S53. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s^{-1}) in HEL_TR.

Table S20. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in LEI_TR. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	SO ₂	NO	NO ₂
1	95	28	34	18	2	28
2	291 (25)	27	36	14	6	8
3	14	12	5	15	37	24
4	39	18	12	12	13	30
UNDETERMINED		15	13	41	42	9

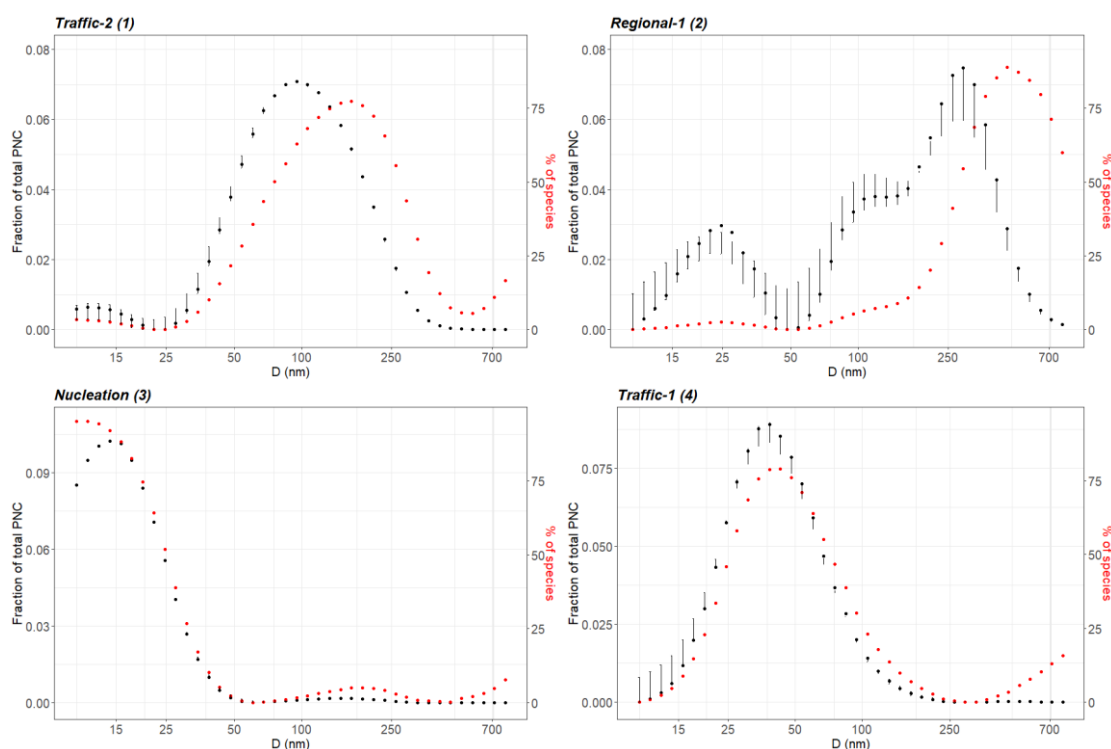


Figure S54. Particle number size distributions of each factor identified by PMF (before splitting) in LEI_TR. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

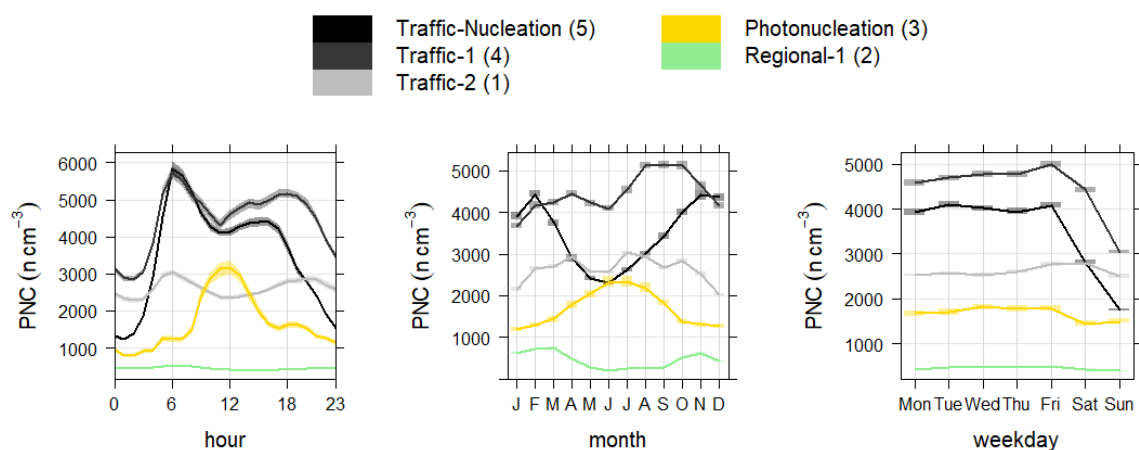


Figure S55. Daily, monthly and weekly patterns of the sources identified (after splitting) in LEI_TR.

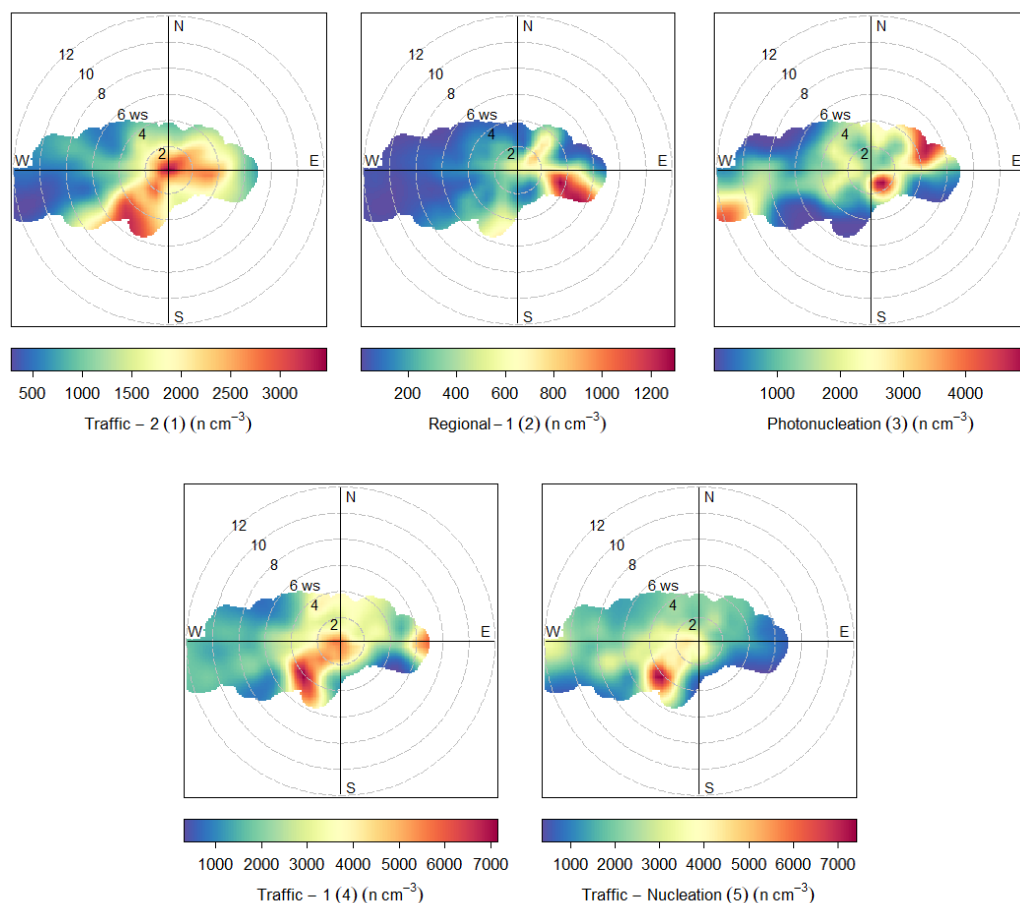


Figure S56. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in LEI_TR.

Table S21. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in LND_TR. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃
1	34	8	25	34	21	45	13	30
2	81	57	24	17	24	26	0	41
3	19	9	0	12	16	11	8	8
4	87	4	40	1	0	9	7	9
UNDETERMINED		22	12	36	39	9	73	12

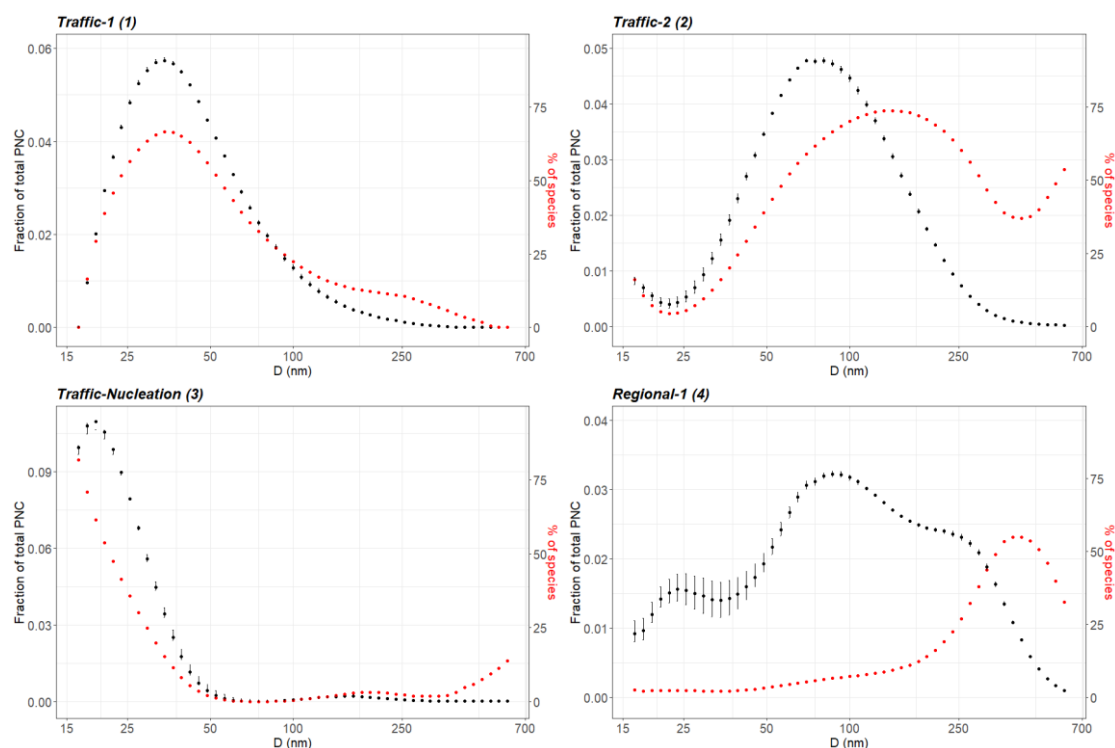


Figure S57. Particle number size distributions of each factor identified by PMF in LND_TR. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

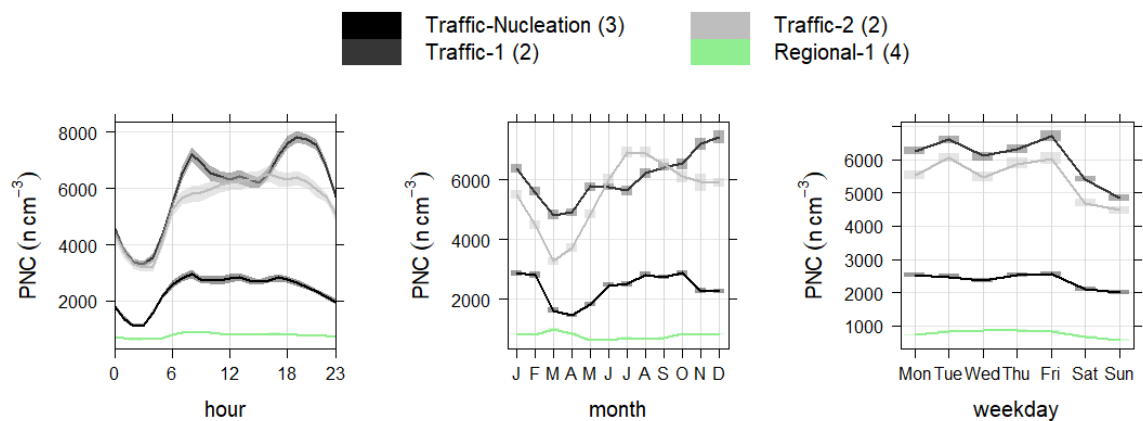


Figure S58. Daily, monthly and weekly patterns of the sources identified in LND_TR.

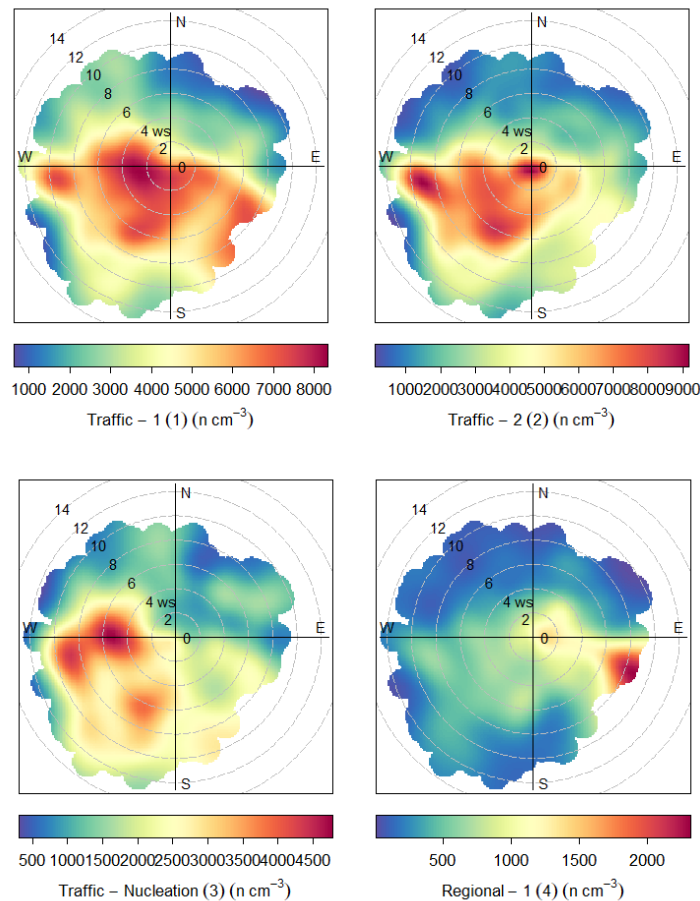


Figure S59. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in LND_TR.

Table S22. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in STO_TR. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	PM _{2.5}	NO	NO ₂	CO
1	17	0	3	2	7	7
2	79	9	7	3	14	21
3	35	9	9	2	15	20
4	198	40	72	1	7	22
UNDETERMINED		43	9	91	57	29

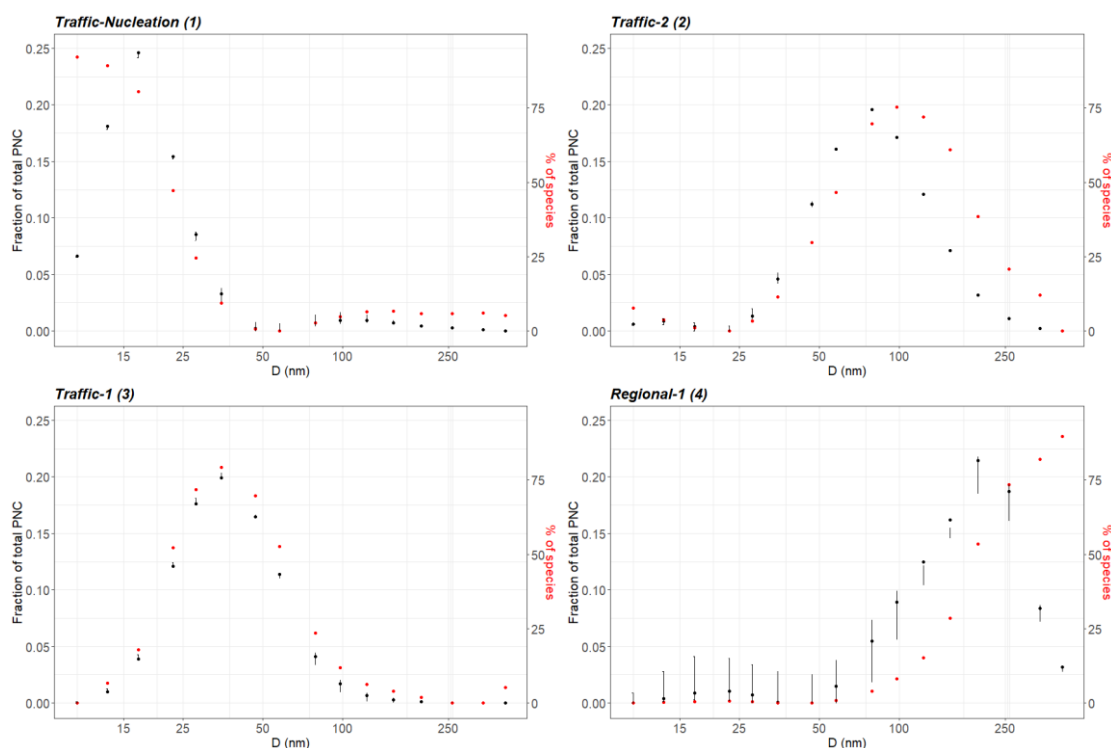


Figure S60. Particle number size distributions of each factor identified by PMF in STO_TR. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

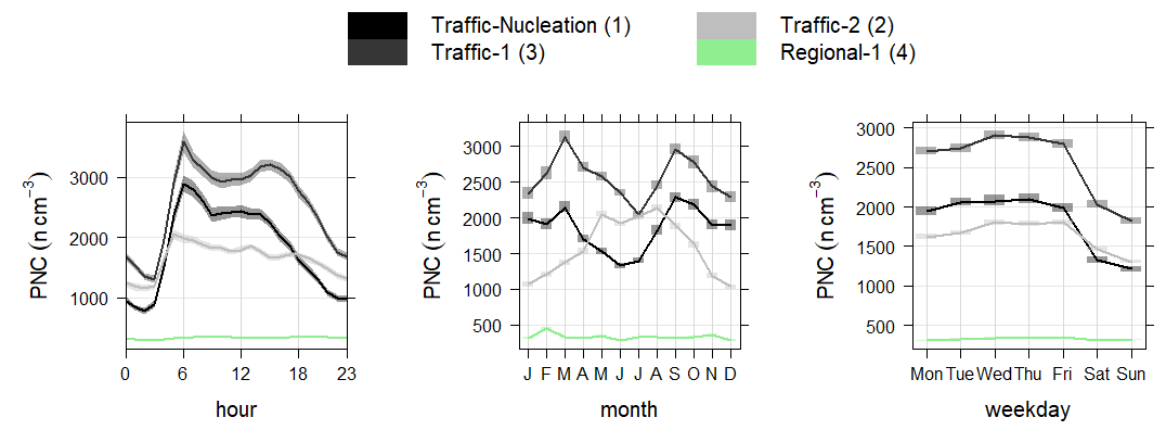


Figure S61. Daily, monthly and weekly patterns of the sources identified in STO_TR.

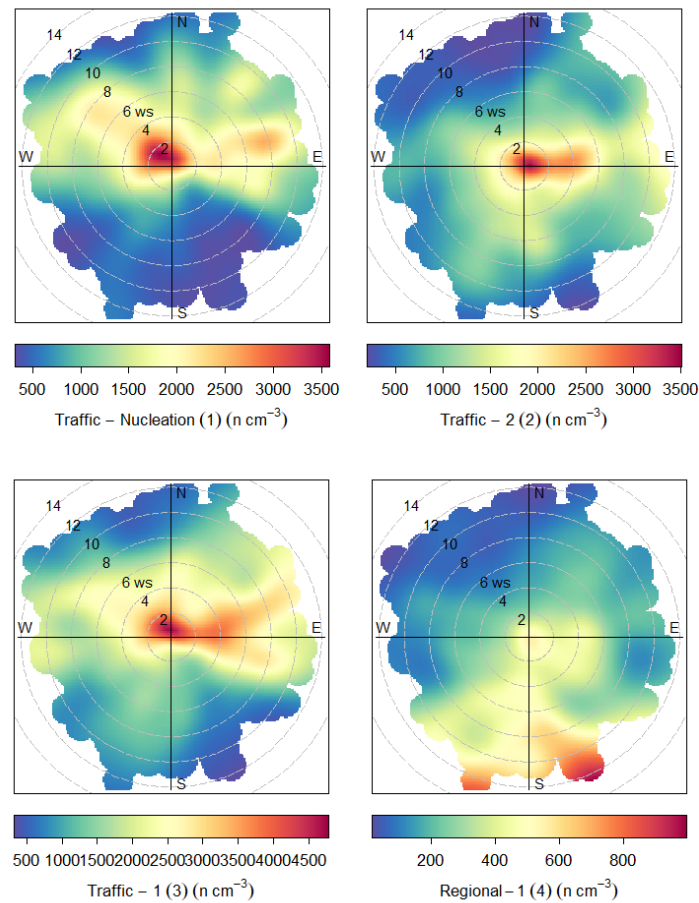


Figure S62. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in STO_TR.

Table S23. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in ATH_SUB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	NO	NO ₂	O ₃
1	70	11	18	25	0
2	178 & 25	55	35	19	59
3	12	1	5	4	0
4	34	6	13	21	14
UNDETERMINED		27	30	32	28

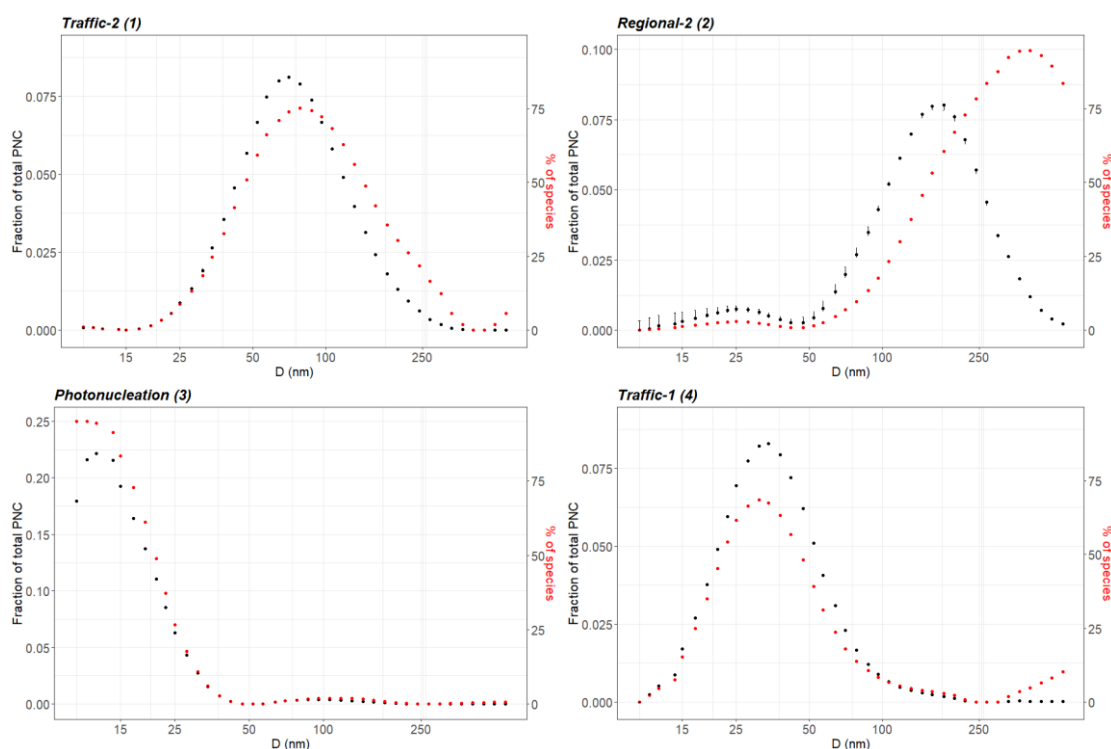


Figure S63. Particle number size distributions of each factor identified by PMF (before splitting) in ATH_SUB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

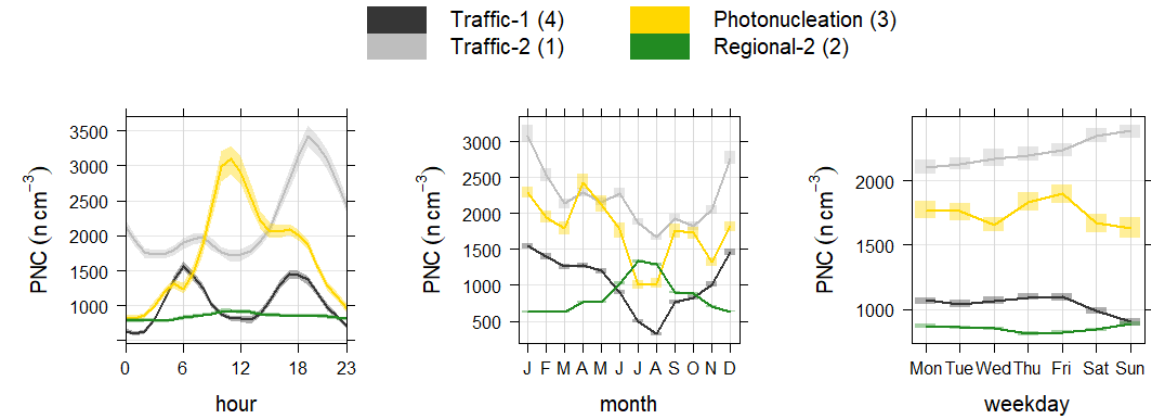


Figure S64. Daily, monthly and weekly patterns of the sources identified (after splitting) in ATH_SUB.

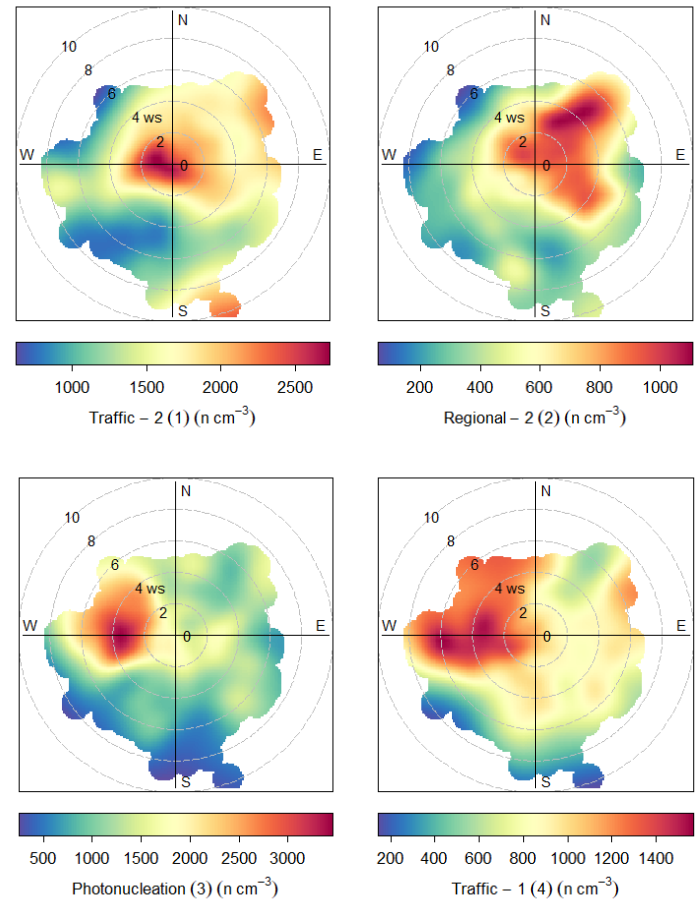


Figure S65. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s⁻¹) in ATH_SUB.

Table S24. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF (before applying splitting methodology) in LIL_SUB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	NO	NO ₂	O ₃
1	106	36	7	33	0
2	24	1	1	1	0
3	269 & 74	18	3	7	0
4	46 & 188	16	2	9	52
5	51	10	5	26	0
UNDETERMINED		18	82	25	48

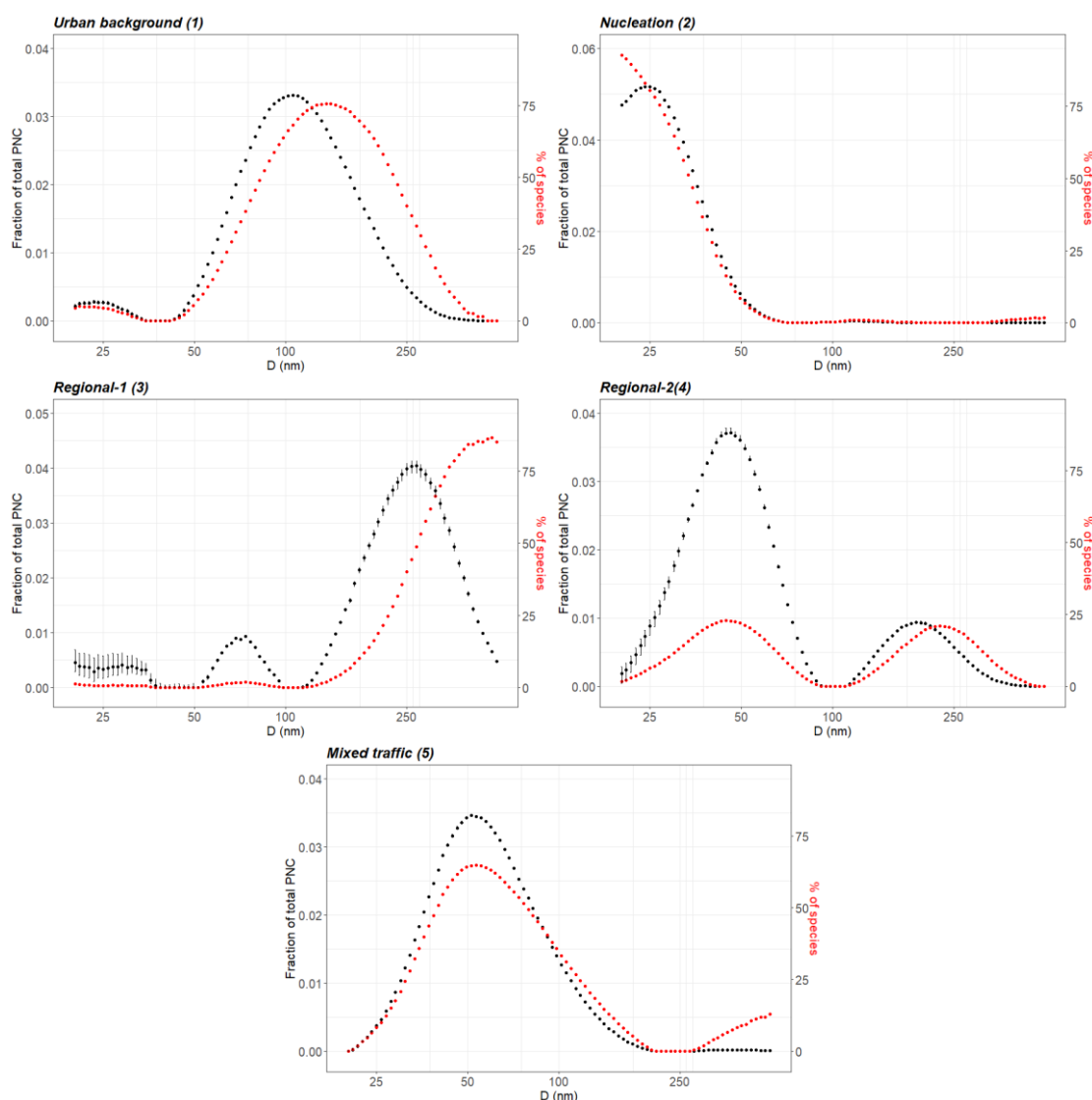


Figure S66. Particle number size distributions of each factor identified by PMF (before splitting) in LIL_SUB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

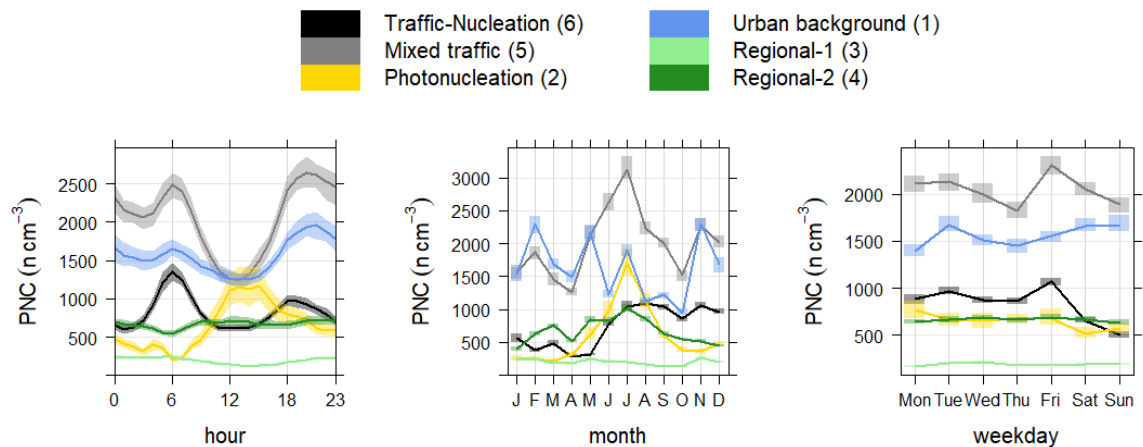


Figure S67. Daily, monthly and weekly patterns of the sources identified (after splitting) in LIL_SUB.

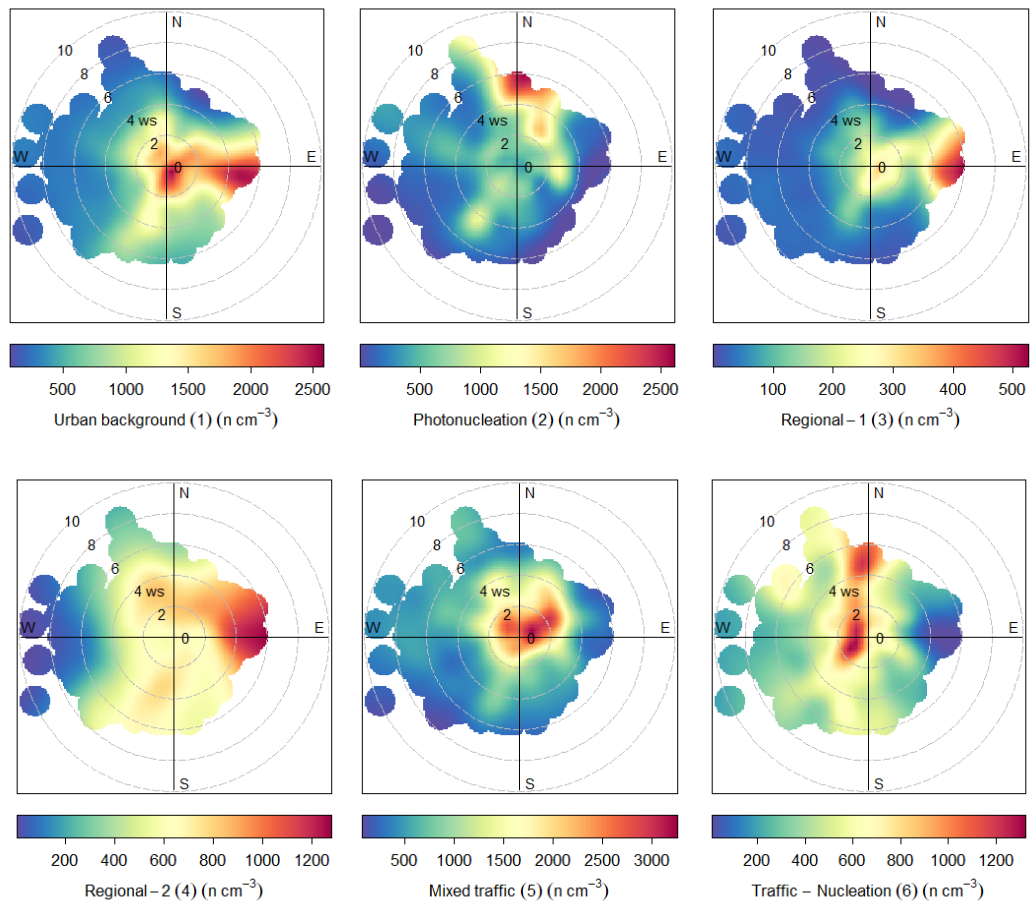


Figure S68. Polar plots showing how contributions of the sources identified (after splitting) are affected by wind direction and wind speed (m s^{-1}) in LIL_SUB.

Table S25. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in PAR_SUB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	PM ₁₀	PM _{2.5}	NO	NO ₂	CO
1	28	10	0	3	11	0
2	199	30	38	2	23	0
3	104	4	9	0	0	78
4	74	36	22	3	20	4
UNDETERMINED		21	32	91	46	18

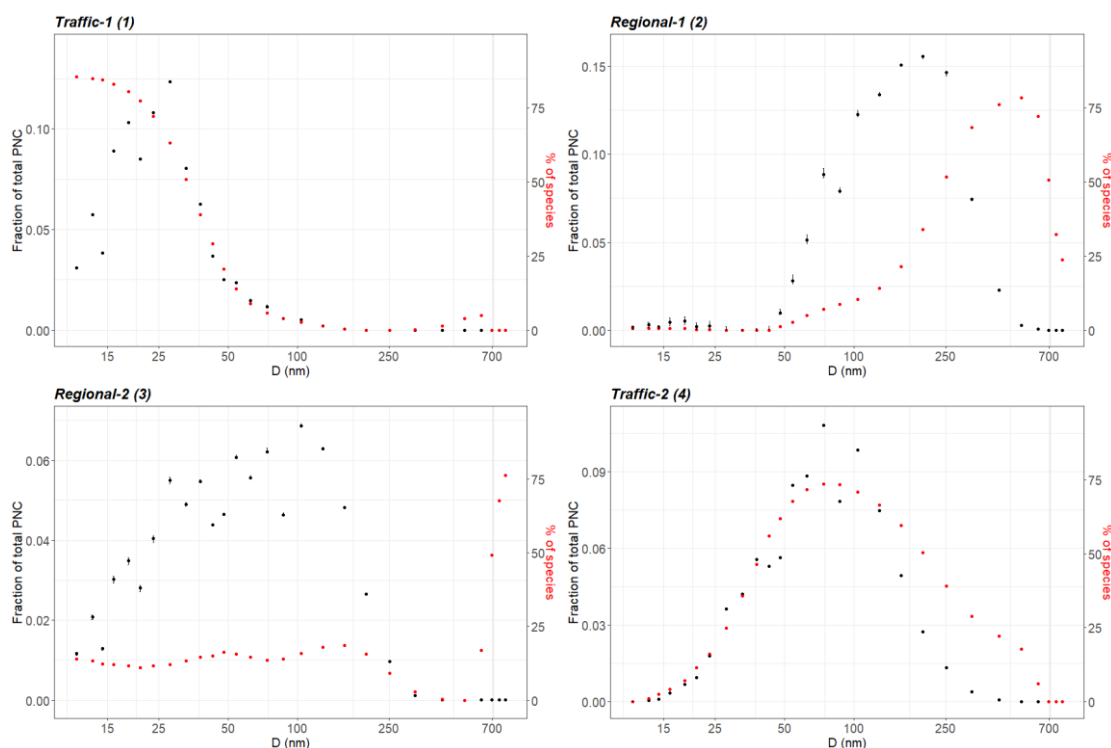


Figure S69. Particle number size distributions of each factor identified by PMF in PAR_SUB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

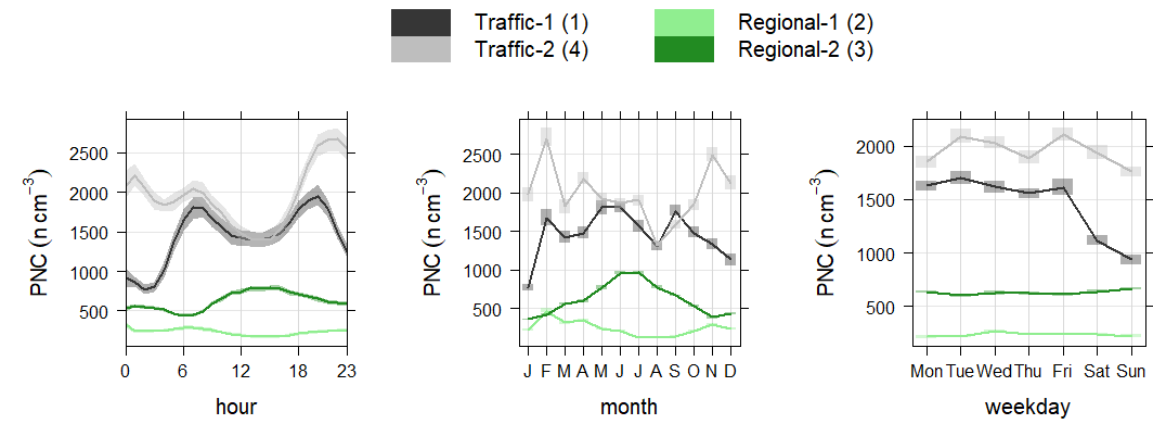


Figure S70. Daily, monthly and weekly patterns of the sources identified in PAR_SUB.

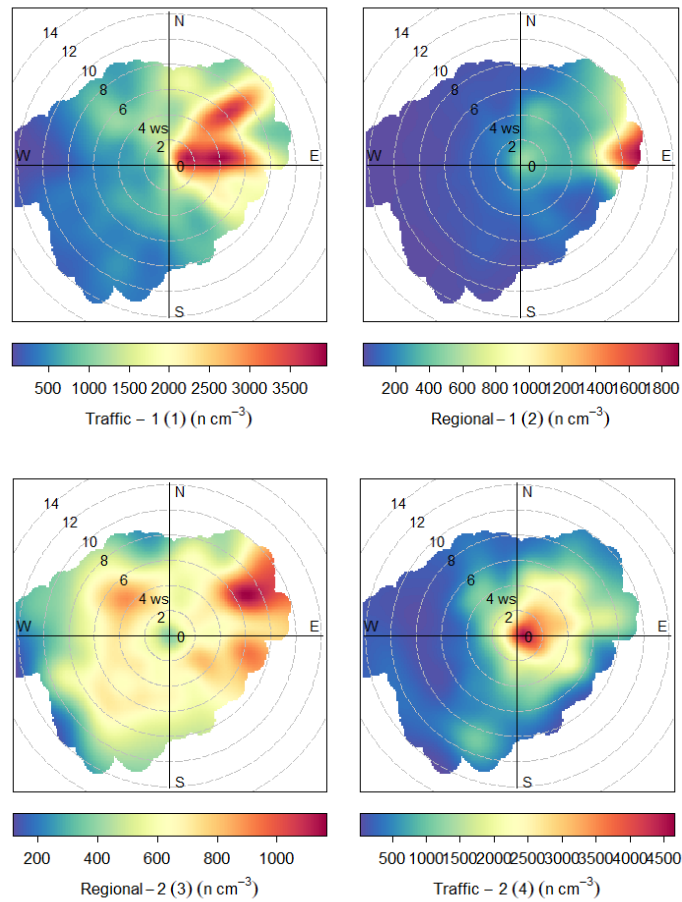


Figure S71. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in PAR_SUB.

Table S26. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in PRA_SUB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃
1	45	10	6	16	3	18	16	18
2	82	7	9	5	3	7	0	13
3	16	6	4	13	3	6	20	12
4	195 & 32	56	58	34	9	39	0	41
UNDETERMINED		22	23	31	83	30	65	16

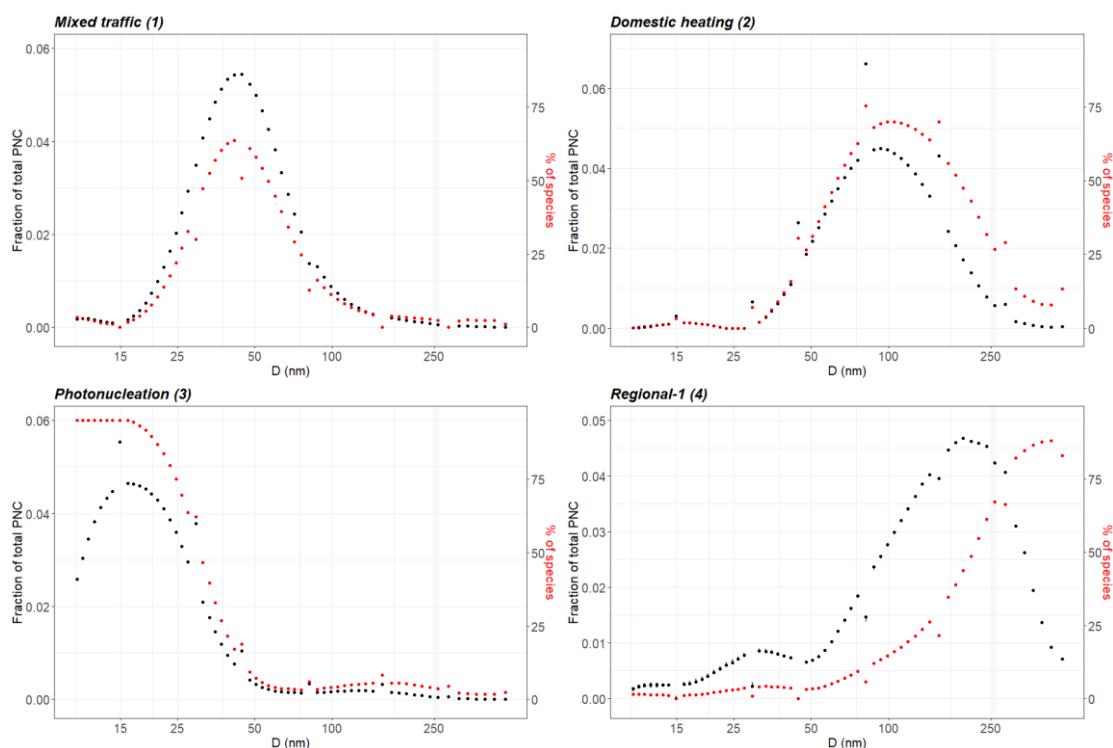


Figure S72. Particle number size distributions of each factor identified by PMF in PRA_SUB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

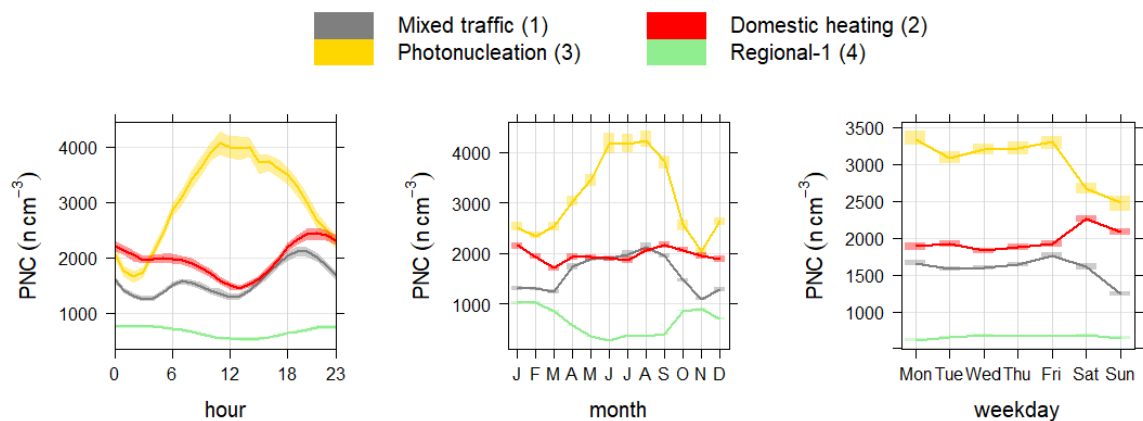


Figure S73. Daily, monthly and weekly patterns of the sources identified in PRA_SUB.

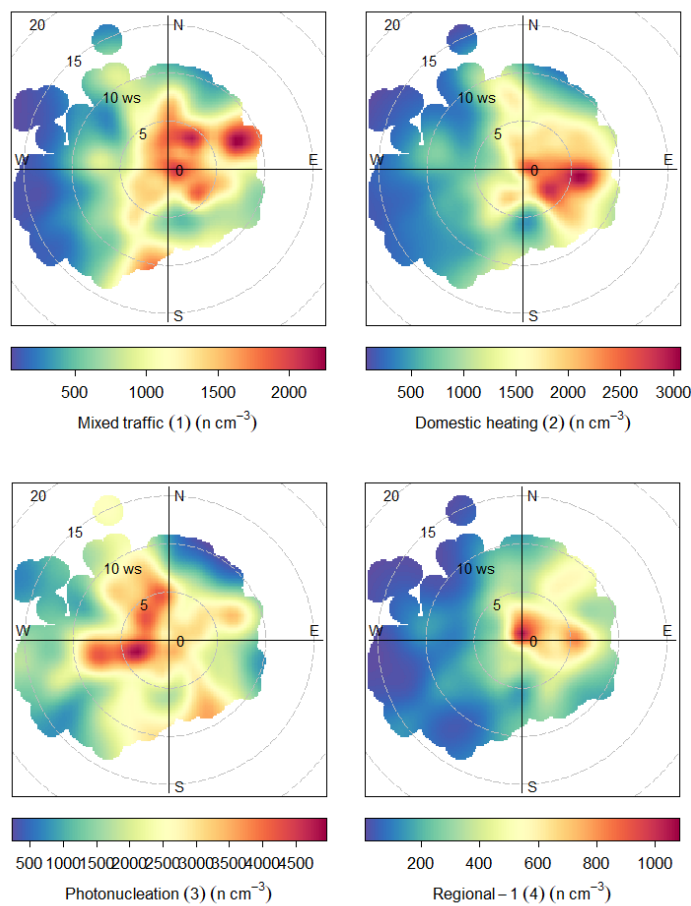


Figure S74. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in PRA_SUB.

Table S27. Size mode and relative contribution (%) of ancillary pollutants of each factor identified by PMF in IPR_RB. UNDETERMINED refers to the relative contributions of ancillary pollutants that have not been attributed to any factor by PMF.

Factor	Mode (nm)	BC	PM ₁₀	SO ₂	NO	NO ₂	O ₃	CO
1	45	13	3	8	0	20	0	14
2	99	45	24	13	1	17	0	29
3	198	31	42	8	1	16	0	11
4	18	1	0	7	1	1	6	4
5	36 & 198	0	17	14	0	2	66	3
UNDETERMINED		10	13	50	96	44	28	40

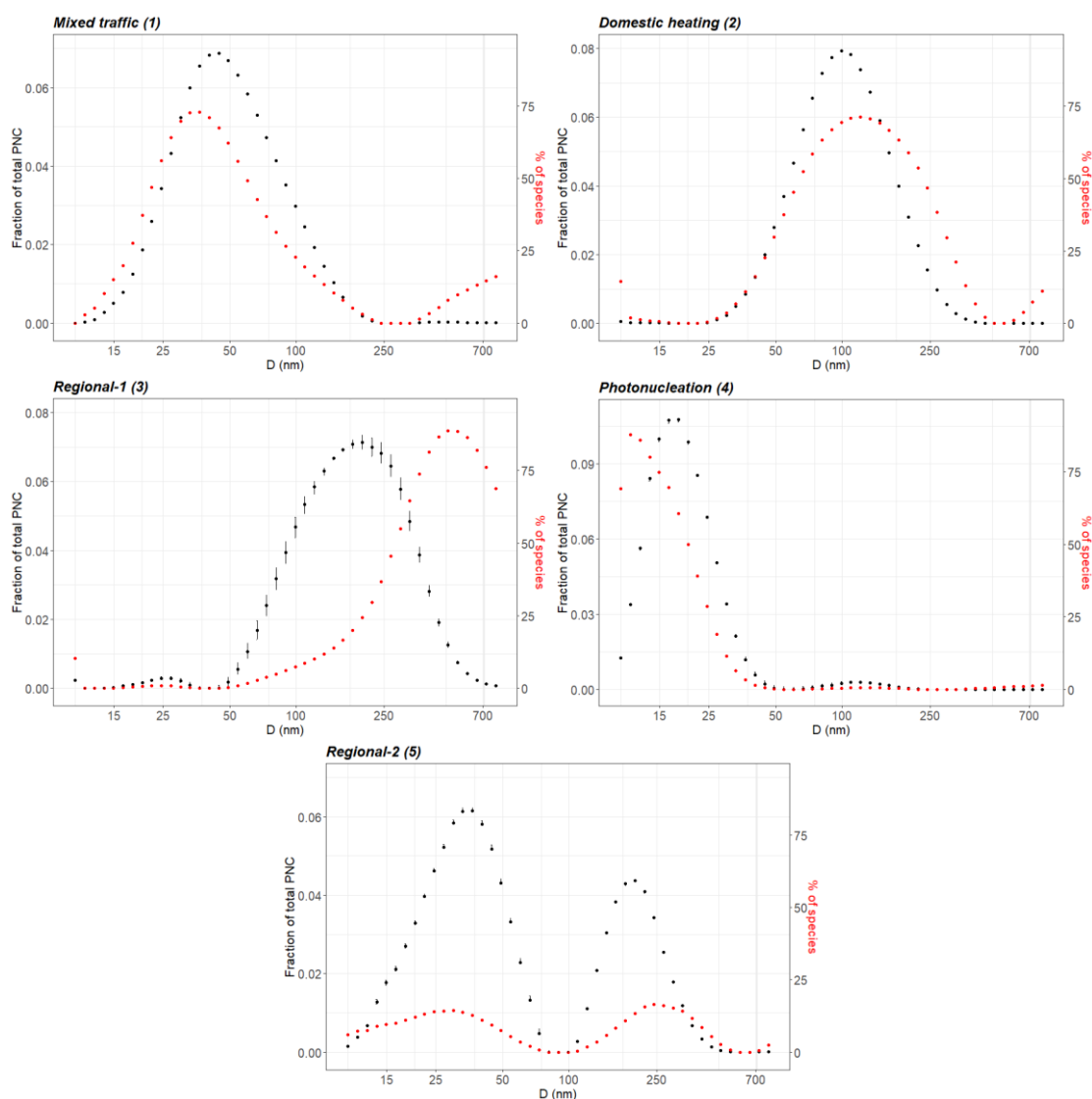


Figure S75. Particle number size distributions of each factor identified by PMF in IPR_RB. The black points represent the normalised fraction of total PNC, the error bars are the minimum and maximum fractional DISP values, and the red points present the % explained variation.

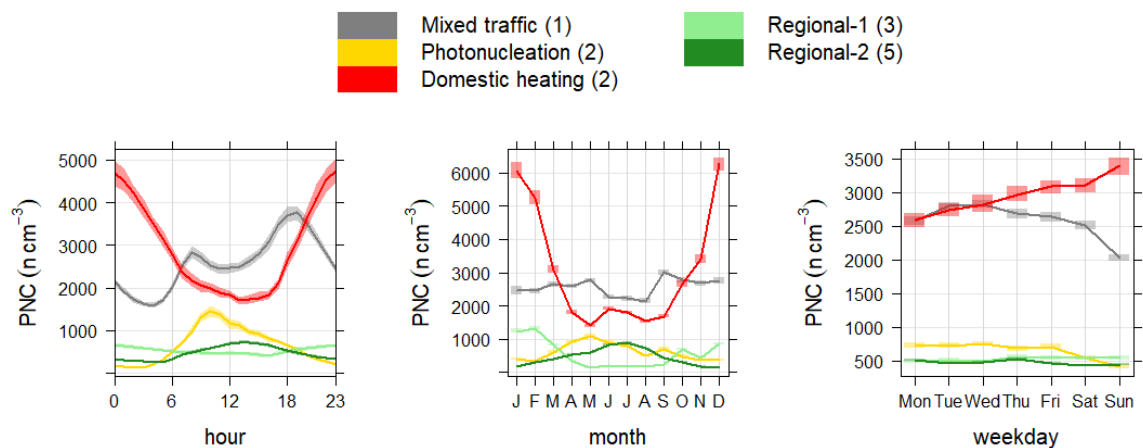


Figure S76. Daily, monthly and weekly patterns of the sources identified in IPR_RB.

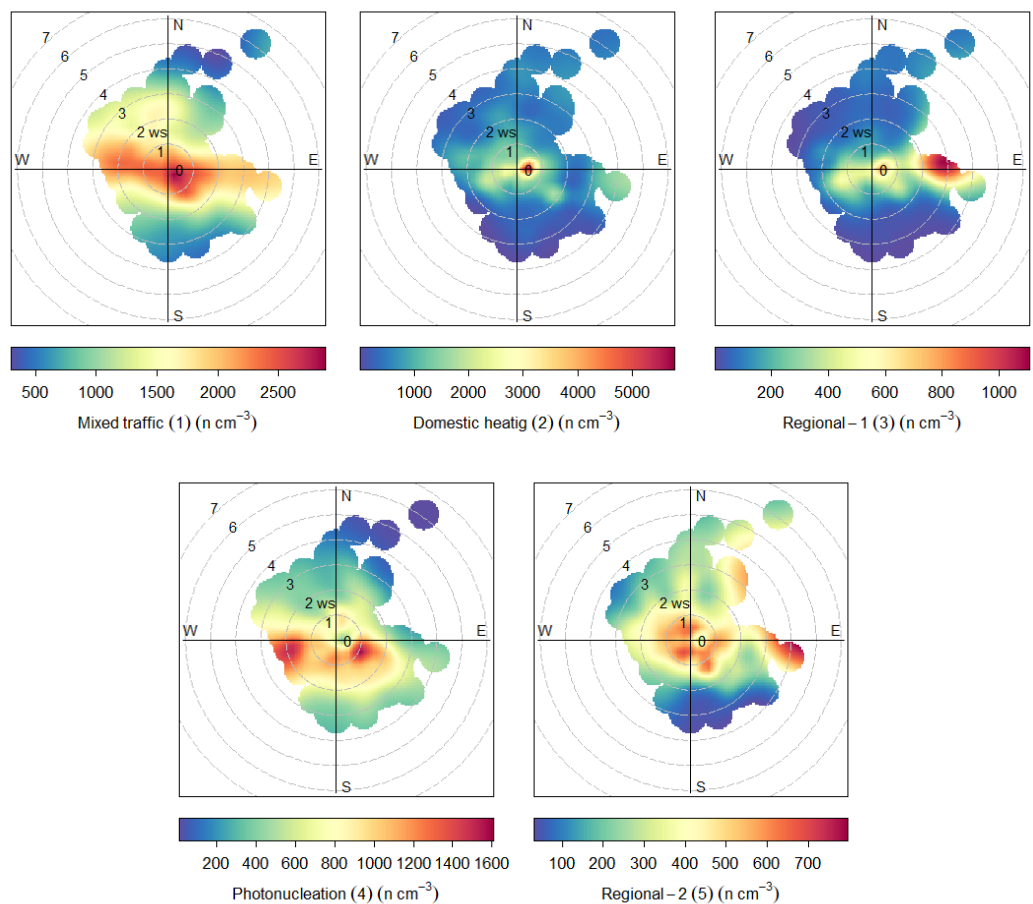
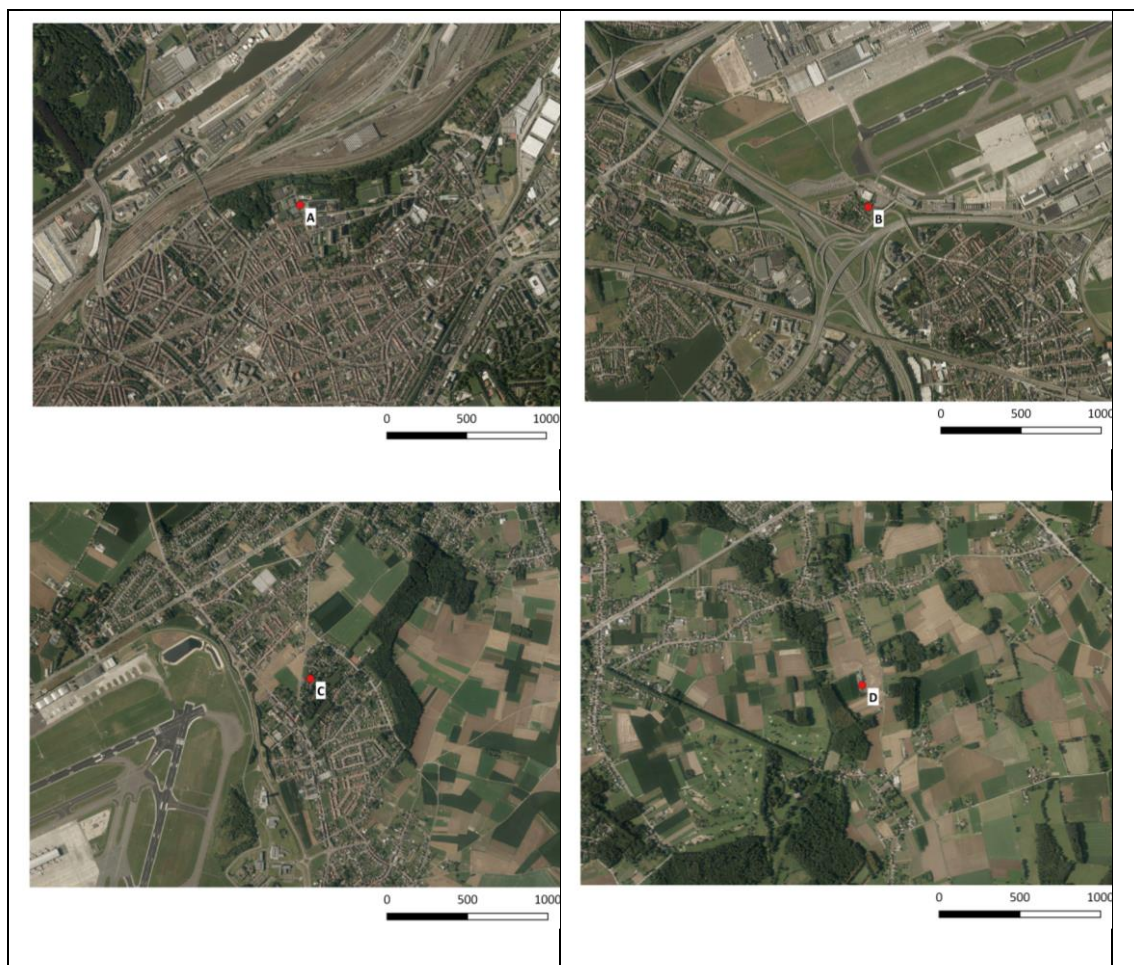


Figure S77. Polar plots showing how contributions of the sources identified are affected by wind direction and wind speed (m s⁻¹) in IPR_RB.

Supplement of paper IV

Spatiotemporal variation and source apportionment of ultrafine particles near Brussels Airport (Belgium)

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S1. Aerial photos of the four monitoring sites.

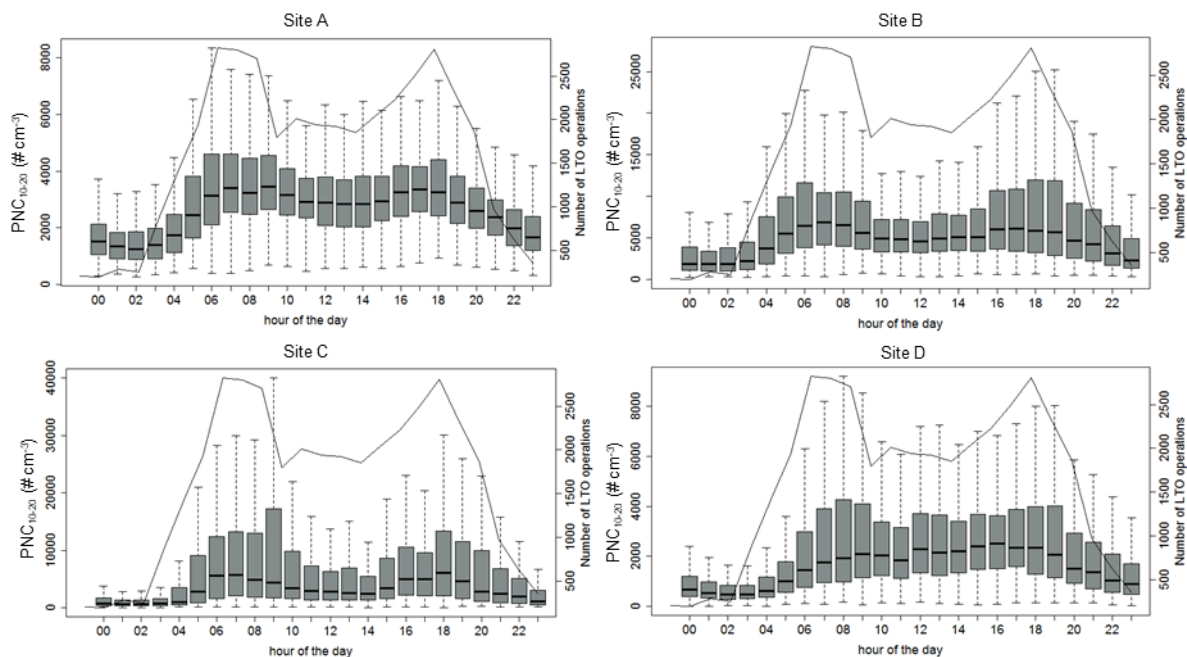
S2: Rescaling factors for the ultrafine particles (UFP) instrumentation using linear regression ($Y = bX$, where Y are the measurements of monitor A). The difference in percentage of UFP number concentrations after rescaling the results of monitors B-D using monitor A as the reference is given in the bottom table.

Particle size class	Rescaling factor (b)			
	Monitor A	Monitor B	Monitor C	Monitor D
10 – 20 nm	1.00	1.00	0.86	0.76
20 – 30 nm	1.00	1.00	0.85	1.00
30 – 50 nm	1.00	0.96	0.79	0.91
50 – 70 nm	1.00	0.91	0.83	0.89
70 – 100 nm	1.00	0.92	0.84	0.90
100 – 200 nm	1.00	0.92	0.78	0.92
200 – 294 nm	1.00	0.93	0.75	0.89

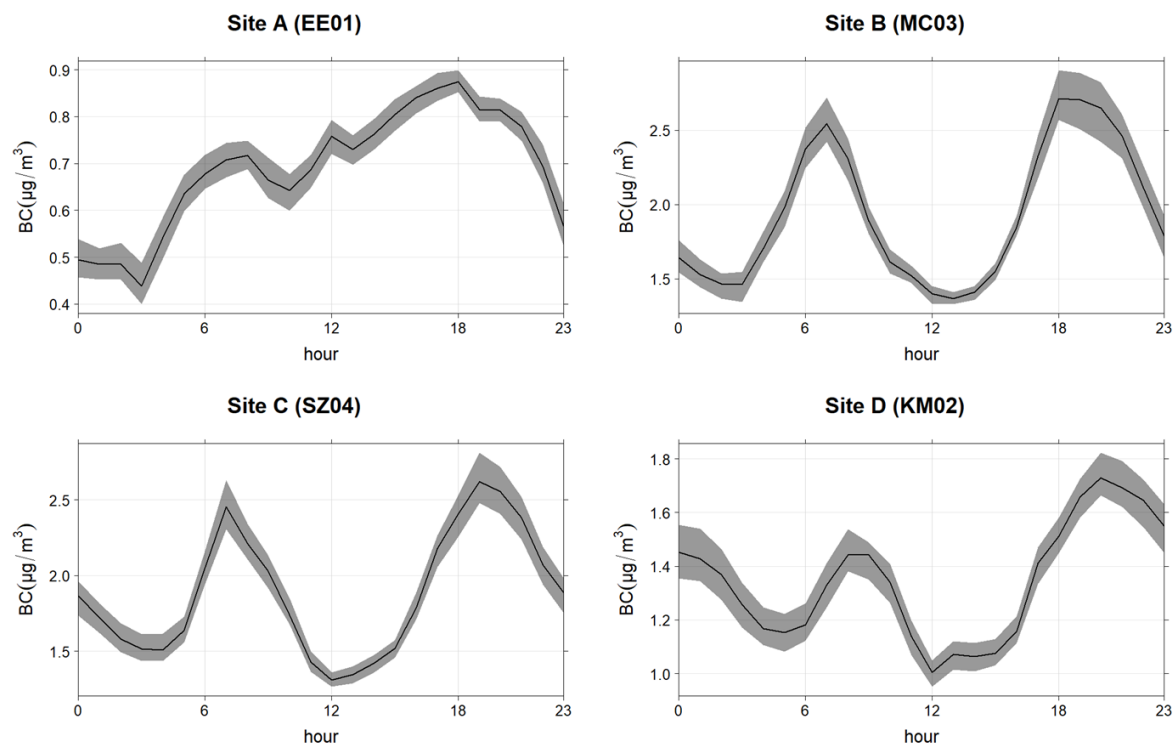
Particle size class	Difference in terms of percentage with Monitor A			
	Monitor A	Monitor B	Monitor C	Monitor D
10 – 20 nm	0%	5%	-8%	0%
20 – 30 nm	0%	8%	-8%	-2%
30 – 50 nm	0%	2%	-4%	-3%
50 – 70 nm	0%	-1%	-3%	1%
70 – 100 nm	0%	0%	-2%	2%
100 – 200 nm	0%	0%	-1%	0%
200 – 294 nm	0%	0%	-2%	-1%

S3: Comparison of meteorological variables in October and November for 2013-2015.

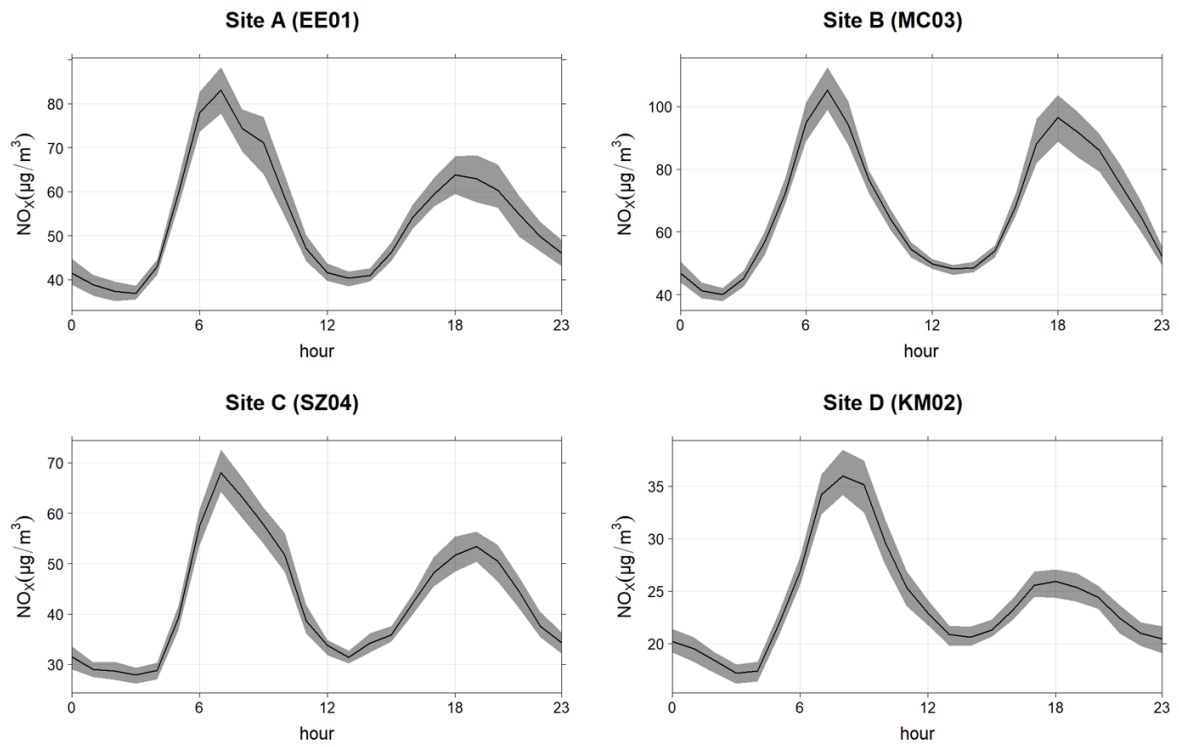
Variable	2013	2014	2015
Temperature	9.9 °C	10.7 °C	10.2 °C
Relative humidity	89 %	90 %	89 %
Rainfall	180 mm	99 mm	145 mm
Precipitation days	41 days	27 days	35 days
Wind speed	3.4 m s ⁻¹	3.2 m s ⁻¹	3.3 m s ⁻¹
Wind direction (proportional, %)			
0 – 90° (N to E)	12 %	13 %	16 %
90 – 180° (E to S)	22 %	58 %	22 %
180 – 270° (S to W)	52 %	27 %	54 %
270 – 360° (W to N)	14 %	2 %	8 %



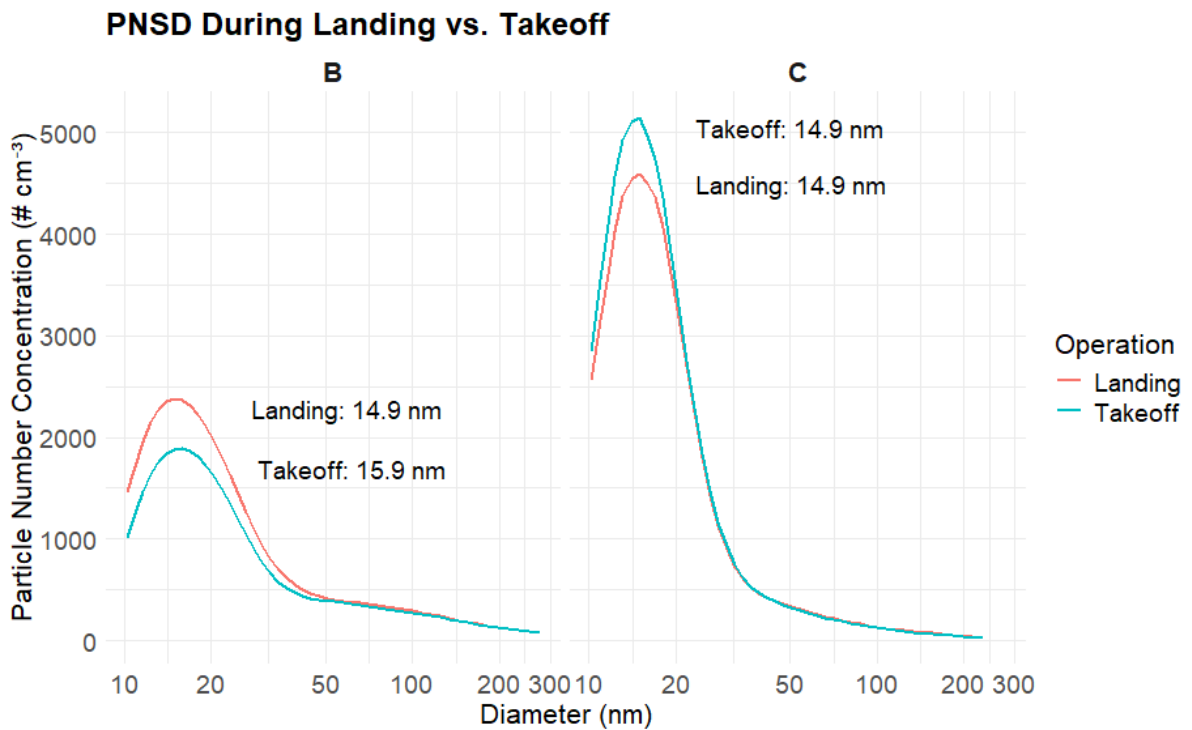
S4: Daily pattern of the 10-20 nm particle number concentration (PNC) at the four sites and the number of Landing and Take-off (LTO) operations at the airport (aggregated over the two-month measurement campaign).



S5: Daily pattern of BC (µg m⁻³) at the four sites.



S6: Daily pattern of NO_x ($\mu\text{g m}^{-3}$) at the four sites.



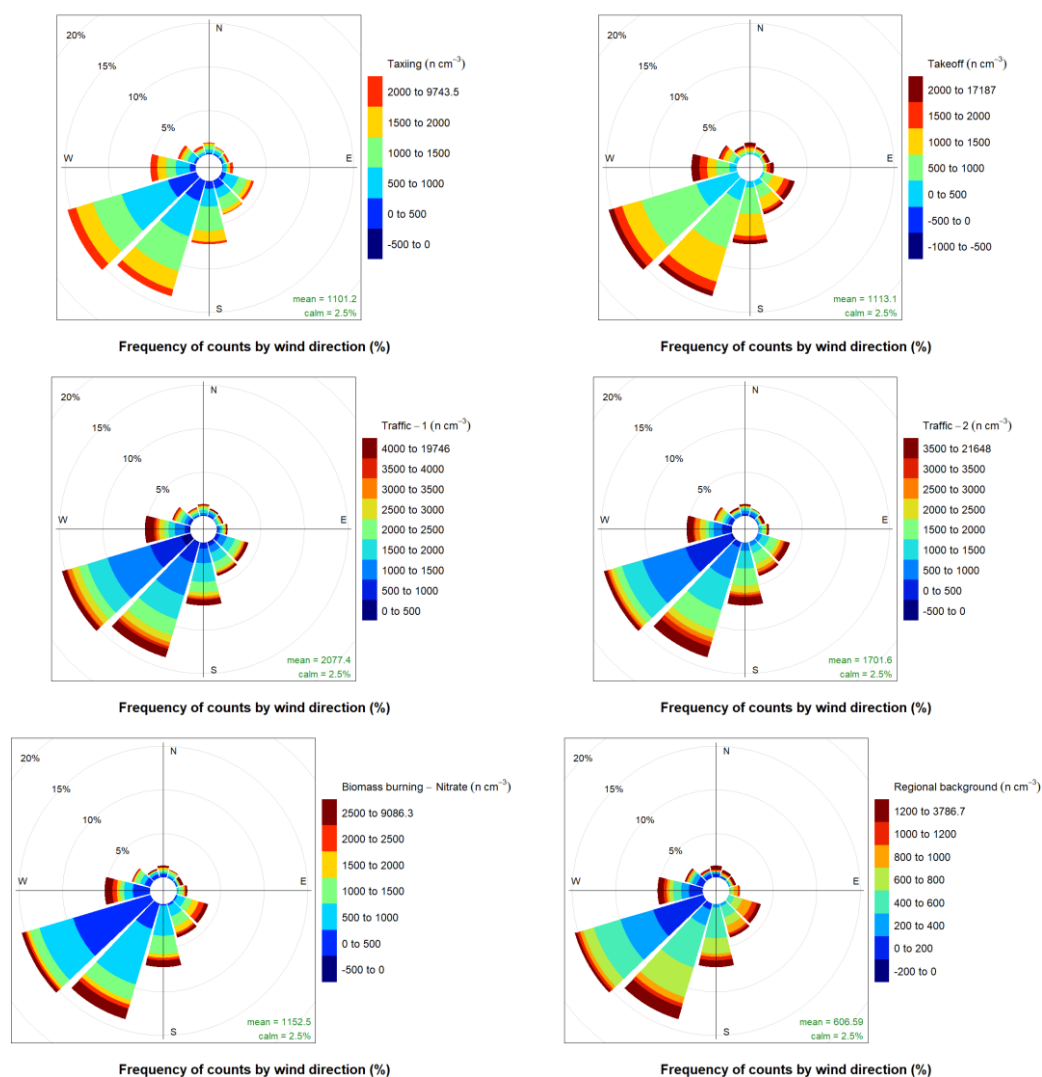
S7: Mean particle number size distributions (PNSD) during landing and takeoff at sites B (left) and C (right). Peak size bins are annotated with their corresponding size (nm).

S8: Size mode (nm) and relative contribution (%) to ancillary pollutants of the six sources identified. UNDETERMINED refers to the relative contributions to ancillary pollutants that have not been attributed to any factor by Positive Matrix Factorization (PMF). Cell color indicates the relative magnitude of the contribution: red (> 30%) = high, orange (10-30%) = moderate, green (5-10%) = low, and blue (< 5%) = very low.

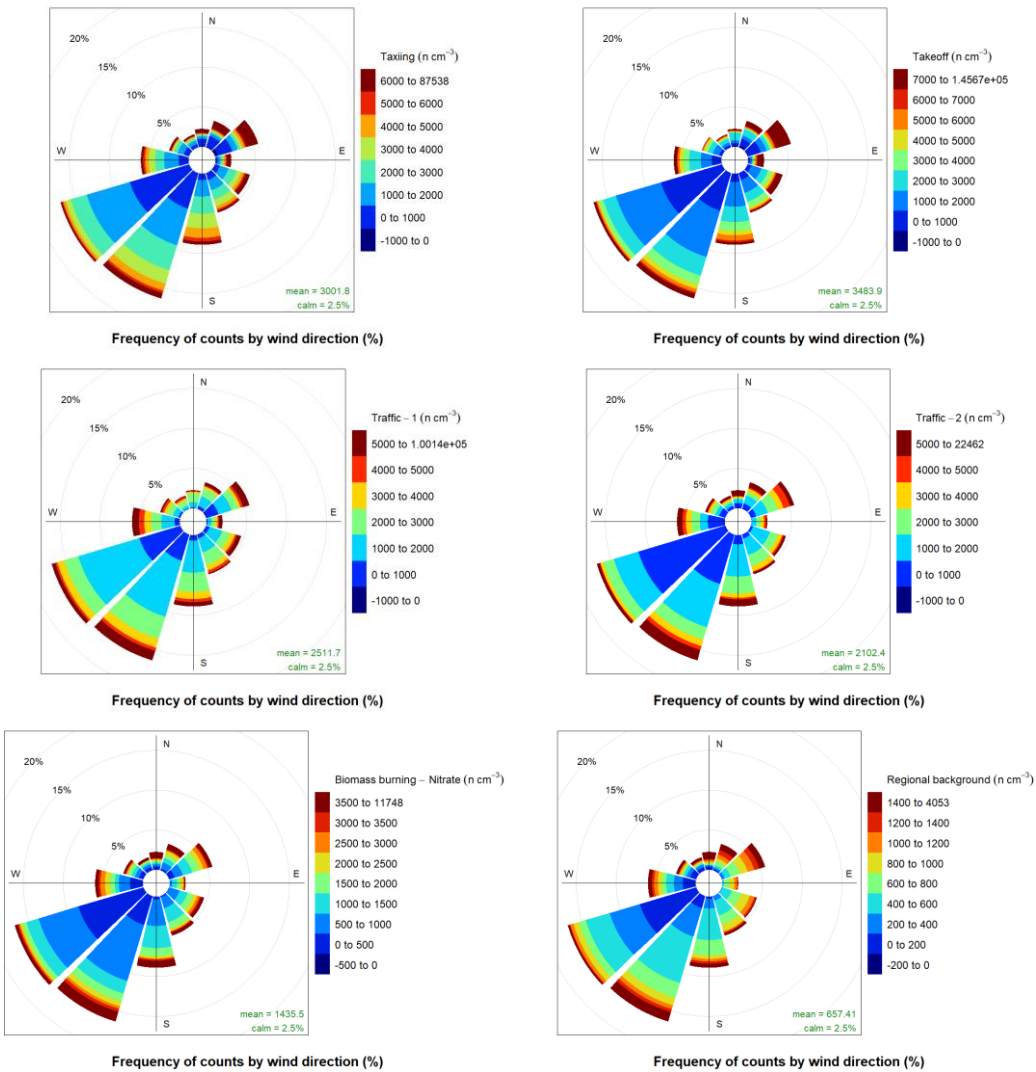
Factor	Mode (nm)	BC	NO	NO ₂
Taxiing	12	1	1	4
Takeoff	20	0	0	0
Traffic-1	32	12	9	39
Traffic-2	62	6	0	11
Biomass burning - Nitrate	126	40	17	5
Regional background	252	25	0	21
UNDETERMINED		16	73	19

S9: Pollution rose plots of the six identified sources at each study location (sites A, B, C, D), showing the frequency of counts by wind direction and wind speed. In green letters: mean indicates the average particle number concentration (PNC, # cm^{-3}) and calm refers to the percentage of time when wind speed was below 0 m s^{-1} .

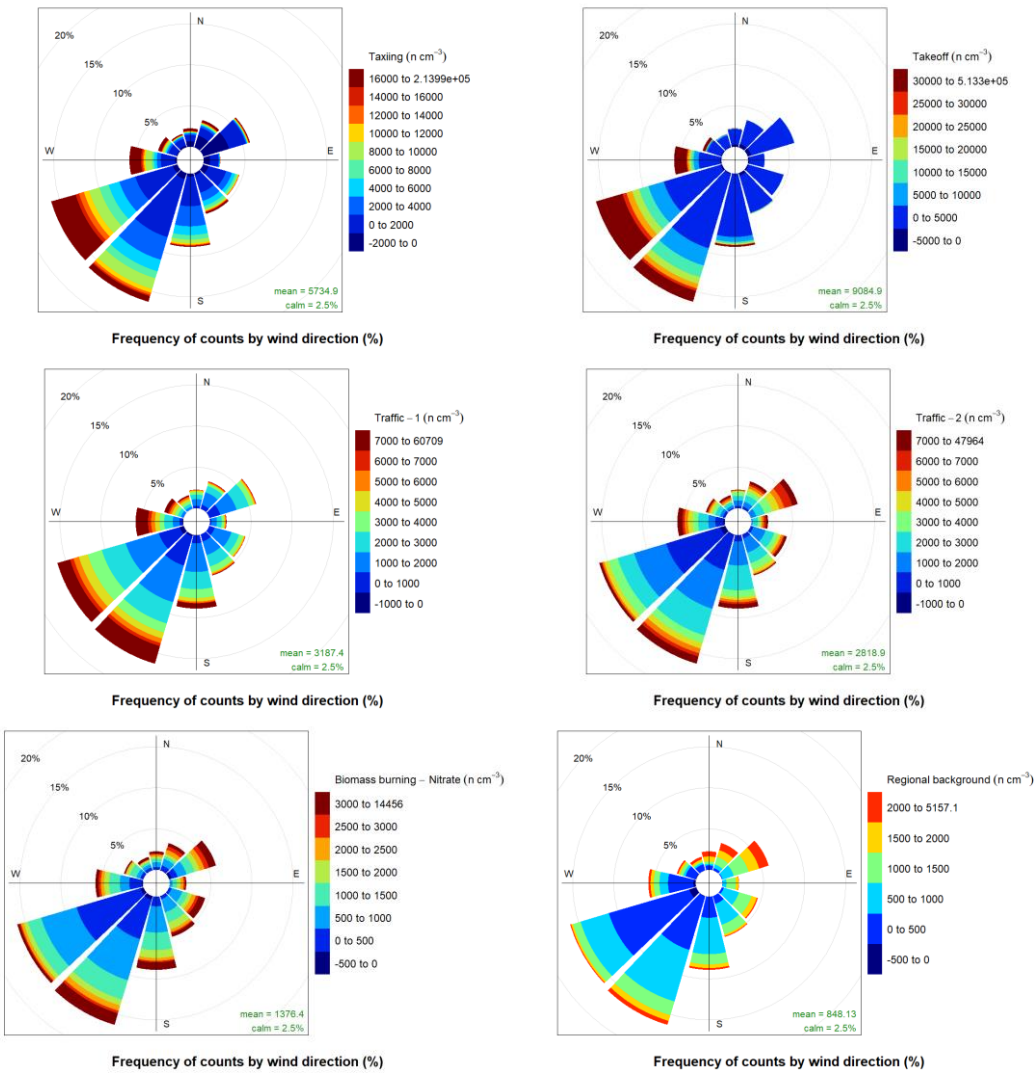
SITE A



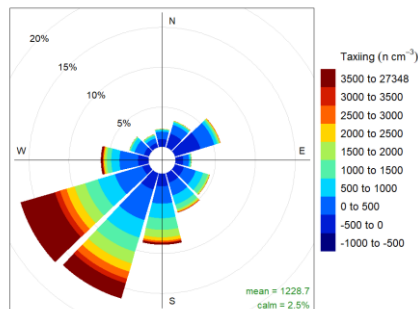
SITE B



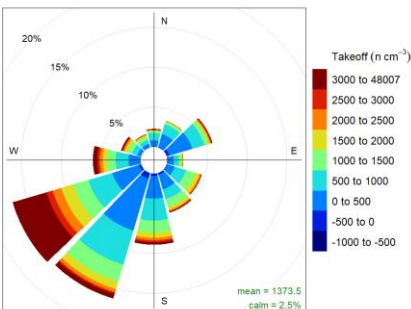
SITE C



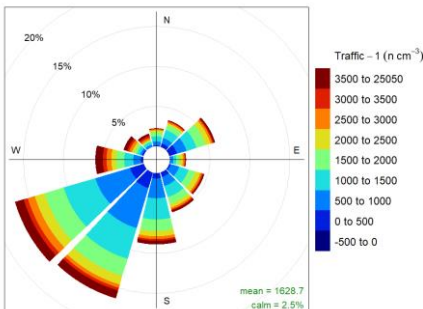
SITE D



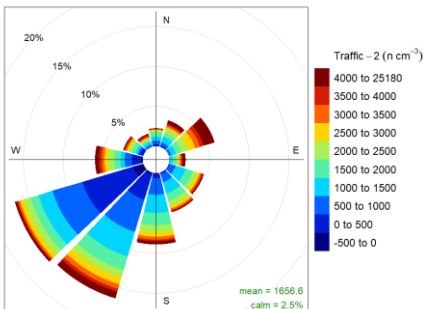
Frequency of counts by wind direction (%)



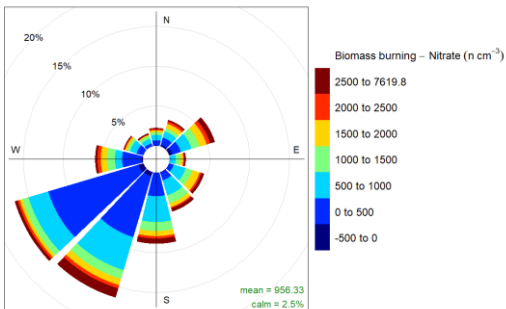
Frequency of counts by wind direction (%)



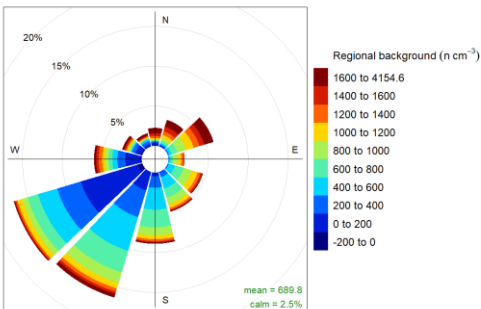
Frequency of counts by wind direction (%)



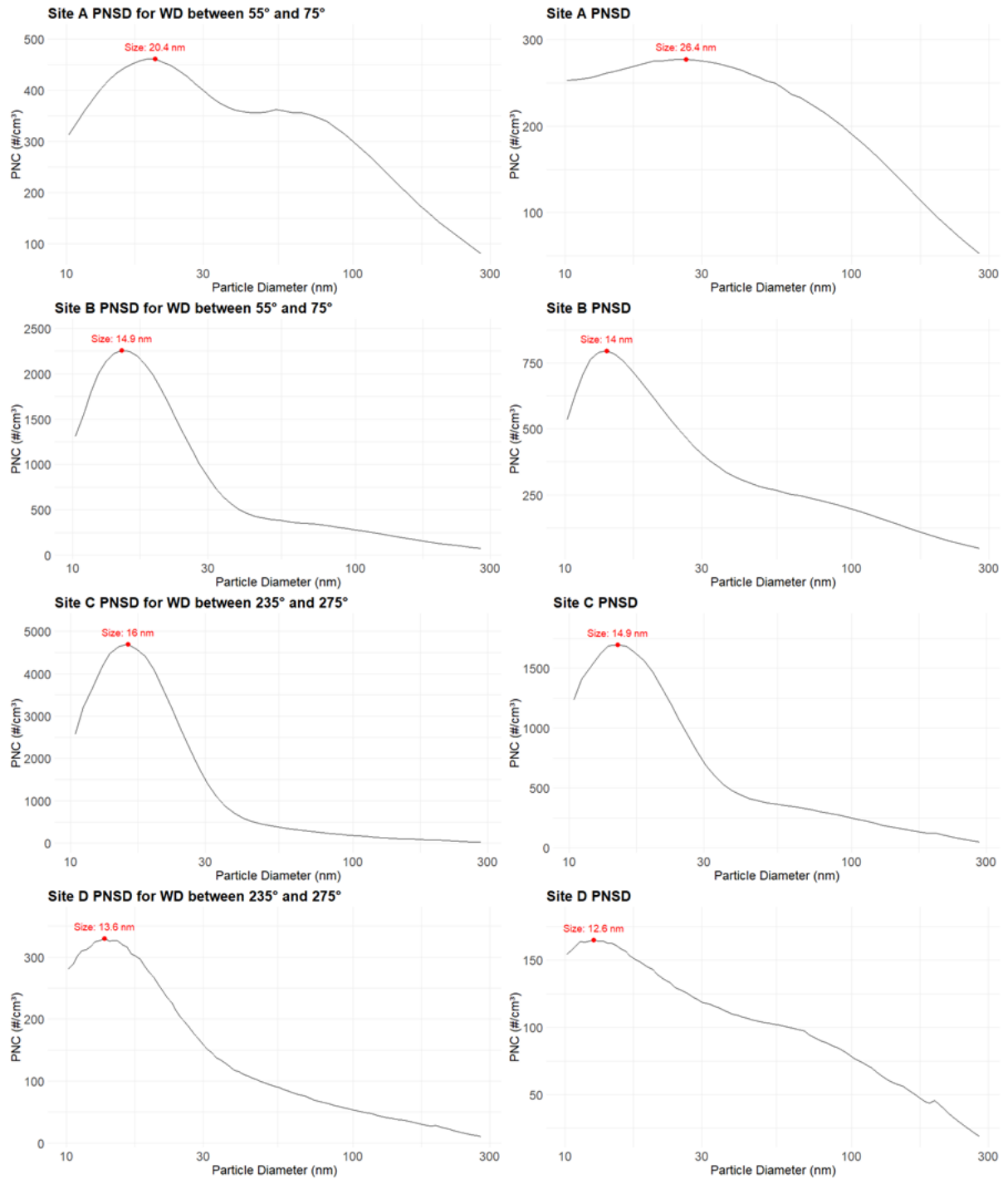
Frequency of counts by wind direction (%)



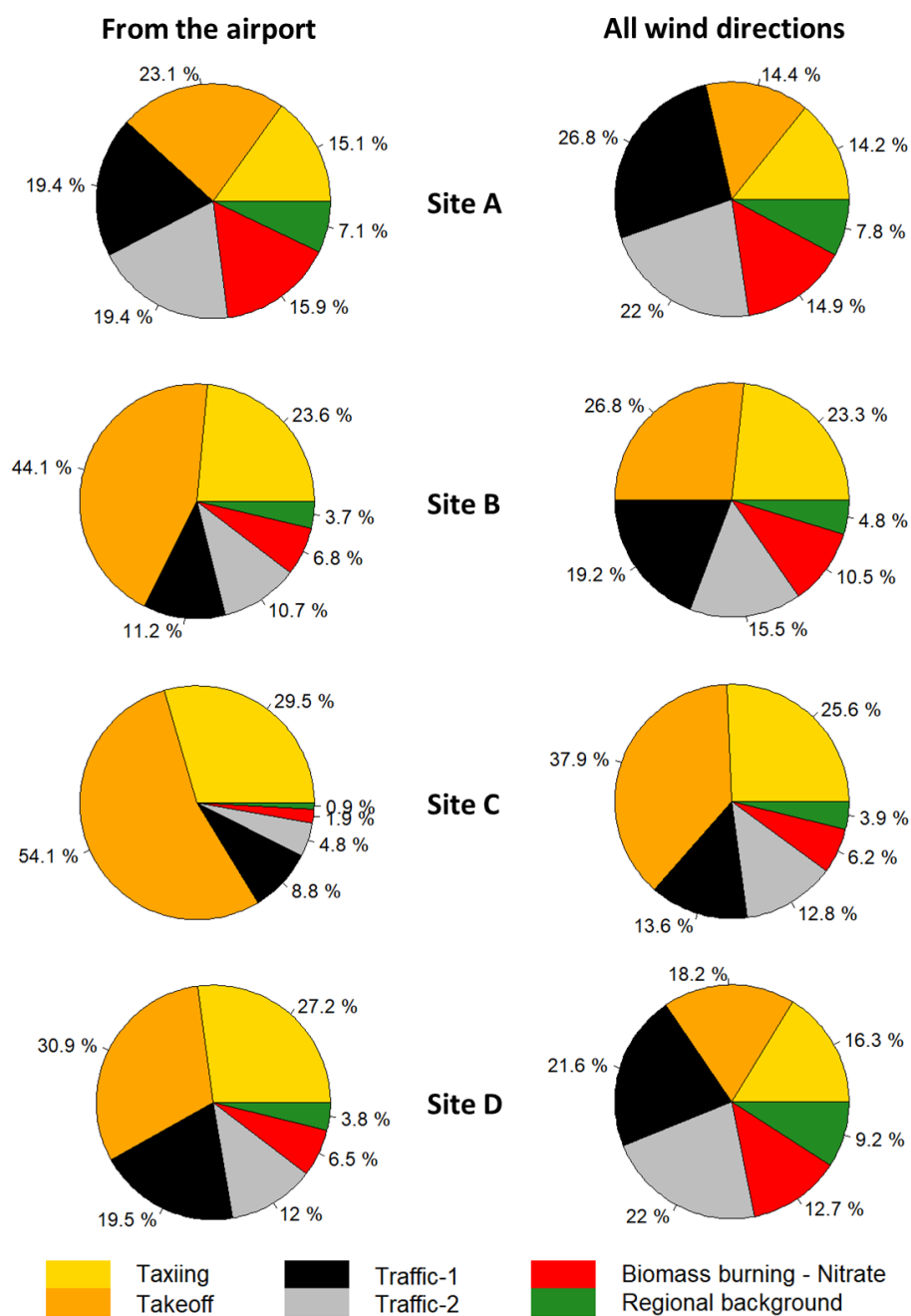
Frequency of counts by wind direction (%)



Frequency of counts by wind direction (%)



S10: Particle number size distributions (PNSDs) at the four monitoring sites, comparing the full dataset (all wind directions, right) with periods when the wind originated from the airport (left). For sites A and B (located southwest of the airport), airport-related wind directions are defined as 55°-75°, and for sites C and D (northeast of the airport), as 235°-275°.



S11: Source contributions to particle number concentration (PNC) identified by Positive Matrix Factorization (PMF) at the four monitoring sites, shown as pie charts for the full dataset (all wind directions) and for subsets corresponding to wind directions from the airport.

Supplement of paper V

Effect of dispersion normalization on long-term source apportionment and trends of ultrafine particles in Barcelona

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Table S1. Selection of the parameters C_3 and K optimising PMF solutions, and obtained $Q_{\text{true}}/Q_{\text{exp}}$ ratio for each dataset analysed.

Dataset	C_3	K	$Q_{\text{true}}/Q_{\text{exp}}$
Dispersion corrected warm season	0.010	0.045	1.09
Uncorrected warm season	0.020	0.050	1.02
Dispersion corrected cold season	0.010	0.040	1.01
Uncorrected cold season	0.020	0.035	1.06

Table S2. Observed (uncorrected) and dispersion normalised (corrected) annual mean particle number concentrations (PNC, in # cm⁻³) for the 12-year study period.

Year	PNC_uncorrected	PNC_corrected
2013	11894	10683
2014	11291	11532
2015	10885	8062
2016	10550	8759
2017	11921	8770
2018	11461	10185
2019	10438	8619
2020	6229	5490
2021	6265	5376
2022	9104	6933
2023	6483	6215
2024	8398	7273

Table S3. Relative contribution (%) to ancillary pollutants of the factors identified by Positive Matrix Factorization (PMF) for the cold season dispersion corrected analysis. “Undetermined” refers to the contributions that have not been attributed to any factor. Cell color indicates the relative magnitude of the contributions: red (> 30%) = high, orange (10-30%) = moderate, green (5-10%) = low, blue (<5%) = very low.

Factor	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃	CO
Traffic-1	4	2	8	8	6	0	12
Mixed traffic	2	0	15	16	30	0	15
Traffic-2	26	30	13	14	37	0	15
Photonucleation	7	5	17	5	1	15	12
Regional-1	6	10	1	0	1	0	1
Regional-2	27	34	17	0	1	23	7
Undetermined	28	19	29	58	24	62	38

Table S4. Relative contribution (%) to ancillary pollutants of the factors identified by Positive Matrix Factorization (PMF) for the warm season dispersion corrected analysis. “Undetermined” refers to the contributions that have not been attributed to any factor. Cell color indicates the relative magnitude of the contributions: red (> 30%) = high, orange (10-30%) = moderate, green (5-10%) = low, blue (<5%) = very low.

Factor	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃	CO
Traffic-1 + Nucleation growth	4	1	7	5	5	0	8
Mixed traffic	10	9	17	9	18	7	21
Traffic-2	15	15	23	17	36	0	14
Photonucleation	3	2	9	6	2	16	4
Regional-1	16	21	0	5	4	0	5
Regional-2	29	33	20	8	7	34	14
Undetermined	24	18	24	49	27	43	34

Table S5. Relative contribution (%) to ancillary pollutants of the factors identified by Positive Matrix Factorization (PMF) for the cold season uncorrected analysis. “Undetermined” refers to the contributions that have not been attributed to any factor. Cell color indicates the relative magnitude of the contributions: red (> 30%) = high, orange (10-30%) = moderate, green (5-10%) = low, blue (<5%) = very low.

Factor	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃	CO
Traffic-1	2	0	4	4	9	0	12
Mixed traffic	0	0	16	4	31	0	19
Traffic-2	40	43	9	6	33	0	18
Photonucleation	5	2	16	3	5	3	9
Regional-1	8	11	7	0	2	0	3
Regional-2	25	28	22	0	0	51	10
Undetermined	20	16	27	83	21	46	30

Table S6. Relative contribution (%) to ancillary pollutants of the factors identified by Positive Matrix Factorization (PMF) for the warm season uncorrected analysis. “Undetermined” refers to the contributions that have not been attributed to any factor. Cell color indicates the relative magnitude of the contributions: red (> 30%) = high, orange (10-30%) = moderate, green (5-10%) = low, blue (<5%) = very low.

Factor	PM ₁₀	PM _{2.5}	SO ₂	NO	NO ₂	O ₃	CO
Traffic-1 + Nucleation growth	4	2	5	1	7	0	8
Mixed traffic	10	10	17	7	22	0	22
Traffic-2	21	21	18	7	36	0	15
Photonucleation	3	1	10	4	3	6	3
Regional-1	18	23	1	3	3	0	3
Regional-2	27	30	24	7	1	60	19
Undetermined	18	14	26	71	28	34	31

Table S7. Summary of Theil-Sen trend analysis results for the full-year, dispersion corrected dataset, including particle number concentration (PNC), size modes, source contributions to PNC, and ancillary pollutants. Reported values are the p-value (p), absolute slope (units per year) with its 95% confidence interval (Lower, Upper), and relative slope expressed as percentage change per year with its 95% confidence interval (Slope percent, Lower percent, Upper percent).

Variable	p	Slope	Lower	Upper	Slope percent	Lower percent	Upper percent
Accumulation	0.000	-50.352	-62.433	-39.962	-4.755	-5.465	-4.170
Aitken	0.000	-248.790	-326.044	-178.729	-5.038	-5.930	-3.967
CO	0.000	-0.009	-0.014	-0.005	-3.690	-5.097	-2.107
Mixed traffic	0.000	-123.439	-158.085	-93.350	-4.227	-4.896	-3.471
NO	0.000	-0.240	-0.311	-0.183	-6.887	-8.021	-5.983
NO ₂	0.000	-0.736	-0.899	-0.604	-5.795	-6.613	-5.255
Nucleation	0.000	-136.873	-223.821	-59.714	-3.386	-4.667	-1.757
O ₃	0.180	-0.499	-1.414	0.185	-1.464	-3.639	0.614
Photonucleation	0.788	-6.877	-54.398	38.826	-0.299	-2.079	1.926
PM ₁₀	0.087	-0.271	-0.669	0.046	-1.606	-3.379	0.298
PM _{2.5}	0.007	-0.213	-0.392	-0.060	-2.118	-3.547	-0.639
PNC	0.000	-439.244	-609.335	-285.175	-4.371	-5.370	-3.127
Regional-1	0.000	-13.053	-20.733	-6.051	-4.254	-5.718	-2.284
Regional-2	0.477	-3.638	-11.363	5.194	-1.057	-2.972	1.965
SO ₂	0.007	-0.017	-0.030	-0.004	-2.437	-4.236	-0.737
Total traffic	0.000	-395.026	-475.967	-309.891	-4.641	-5.116	-3.933
Traffic-1	0.000	-144.121	-182.106	-112.249	-4.711	-5.475	-4.013
Traffic-2	0.000	-125.369	-152.696	-99.039	-5.139	-5.828	-4.416

Table S8. Summary of Theil-Sen trend analysis results for the full-year, uncorrected dataset, including particle number concentration (PNC), size modes, source contributions to PNC, and ancillary pollutants. Reported values are the p-value (p), absolute slope (units per year) with its 95% confidence interval (Lower, Upper), and relative slope expressed as percentage change per year with its 95% confidence interval (Slope percent, Lower percent, Upper percent).

Variable	p	Slope	Lower	Upper	Slope percent	Lower percent	Upper percent
Accumulation	0.000	-68.257	-85.705	-50.606	-4.400	-5.194	-3.445
Aitken	0.000	-282.388	-344.092	-221.981	-4.302	-4.909	-3.708
CO	0.000	-0.010	-0.013	-0.007	-3.451	-4.221	-2.562
Mixed traffic	0.000	-137.998	-166.626	-108.855	-4.635	-5.304	-3.996
NO	0.000	-0.410	-0.543	-0.297	-6.632	-7.490	-5.497
NO ₂	0.000	-0.985	-1.133	-0.831	-5.114	-5.598	-4.526
Nucleation	0.000	-97.957	-153.554	-53.787	-2.452	-3.517	-1.503
O ₃	0.641	0.090	-0.283	0.488	0.327	-0.947	1.916
Photonucleation	0.237	-26.855	-72.688	17.374	-1.007	-2.391	0.740
PM ₁₀	0.220	-0.149	-0.400	0.070	-0.743	-1.857	0.358
PM _{2.5}	0.013	-0.197	-0.344	-0.056	-1.455	-2.395	-0.430
PNC	0.000	-453.608	-567.141	-349.049	-3.743	-4.332	-3.044
Regional-1	0.003	-6.556	-11.070	-2.181	-3.936	-5.636	-1.469
Regional-2	0.287	4.575	-4.282	11.816	1.236	-1.007	3.665
SO ₂	0.043	-0.013	-0.024	0.000	-1.888	-3.256	-0.030
Total traffic	0.000	-400.149	-477.752	-320.434	-4.761	-5.298	-4.173
Traffic-1	0.000	-153.478	-198.946	-112.055	-4.759	-5.695	-3.798
Traffic-2	0.000	-107.646	-134.991	-85.415	-5.042	-5.766	-4.276

Table S9. Summary of Theil-Sen trend analysis results for the cold season, dispersion corrected dataset, including particle number concentration (PNC), size modes, source contributions to PNC, and ancillary pollutants. Reported values are the p-value (p), absolute slope (units per year) with its 95% confidence interval (Lower, Upper), and relative slope expressed as percentage change per year with its 95% confidence interval (Slope percent, Lower percent, Upper percent).

Variable	p	Slope	Lower	Upper	Slope percent	Lower percent	Upper percent
Accumulation	0.000	-51.544	-67.371	-36.853	-5.279	-6.084	-4.115
Aitken	0.000	-269.882	-383.509	-175.333	-5.460	-6.654	-4.084
CO	0.003	-0.011	-0.021	-0.003	-3.729	-6.029	-1.077
Mixed traffic	0.000	-160.817	-215.358	-106.745	-4.451	-5.358	-3.318
NO	0.000	-0.360	-0.457	-0.287	-7.412	-8.363	-6.858
NO ₂	0.000	-0.901	-1.129	-0.718	-6.310	-6.885	-5.520
Nucleation	0.023	-144.734	-300.307	-22.852	-3.450	-5.389	-0.656
O ₃	0.107	-0.399	-1.965	0.542	-1.325	-5.023	2.230
Photonucleation	0.060	35.451	-6.042	84.558	1.905	-0.275	5.407
PM ₁₀	0.010	-0.511	-1.043	0.037	-2.774	-4.727	0.241
PM _{2.5}	0.000	-0.326	-0.633	-0.052	-3.084	-5.158	-0.572
PNC	0.080	-463.894	-745.457	-237.474	-4.613	-6.289	-2.668
Regional-1	0.678	-14.097	-28.984	0.668	-3.895	-6.231	0.234
Regional-2	0.411	-1.915	-9.882	7.492	-0.963	-3.792	4.852
SO ₂	0.000	-0.008	-0.028	0.013	-1.303	-3.736	2.676
Total traffic	0.000	-443.738	-596.868	-305.412	-4.688	-5.725	-3.600
Traffic-1	0.000	-152.809	-201.775	-103.485	-4.634	-5.532	-3.469
Traffic-2	0.000	-119.058	-173.190	-71.951	-4.983	-6.382	-3.342

Table S10. Summary of Theil-Sen trend analysis results for the cold season, uncorrected dataset, including particle number concentration (PNC), size modes, source contributions to PNC, and ancillary pollutants. Reported values are the p-value (p), absolute slope (units per year) with its 95% confidence interval (Lower, Upper), and relative slope expressed as percentage change per year with its 95% confidence interval (Slope percent, Lower percent, Upper percent).

Variable	p	Slope	Lower	Upper	Slope percent	Lower percent	Upper percent
Accumulation	0.000	-66.780	-96.541	-31.620	-4.394	-5.848	-2.411
Aitken	0.000	-288.163	-389.586	-201.632	-4.319	-5.311	-3.298
CO	0.000	-0.009	-0.013	-0.004	-2.838	-3.839	-1.472
Mixed traffic	0.000	-160.847	-211.189	-108.228	-5.040	-6.025	-3.910
NO	0.000	-0.650	-0.906	-0.447	-6.568	-7.435	-5.528
NO ₂	0.000	-0.944	-1.201	-0.730	-4.775	-5.722	-3.908
Nucleation	0.003	-73.388	-120.454	-26.990	-1.895	-2.829	-0.784
O ₃	0.337	0.145	-0.206	0.494	0.742	-0.958	2.785
Photonucleation	0.661	13.486	-36.486	50.372	0.563	-1.336	2.353
PM ₁₀	0.164	-0.268	-0.640	0.120	-1.316	-2.728	0.689
PM _{2.5}	0.047	-0.261	-0.512	-0.003	-1.854	-3.189	-0.021
PNC	0.000	-421.717	-588.405	-274.132	-3.446	-4.256	-2.443
Regional-1	0.137	-5.565	-12.662	1.437	-3.669	-6.550	1.308
Regional-2	0.301	6.329	-5.397	17.319	2.099	-1.597	7.433
SO ₂	0.978	0.001	-0.015	0.018	0.090	-2.380	3.788
Total traffic	0.000	-439.588	-582.119	-304.158	-4.830	-5.726	-3.707
Traffic-1	0.000	-171.384	-223.759	-117.112	-4.636	-5.433	-3.391
Traffic-2	0.000	-106.382	-152.199	-63.833	-5.100	-6.507	-3.471

Table S11. Summary of Theil-Sen trend analysis results for the warm season, dispersion corrected dataset, including particle number concentration (PNC), size modes, source contributions to PNC, and ancillary pollutants. Reported values are the p-value (p), absolute slope (units per year) with its 95% confidence interval (Lower, Upper), and relative slope expressed as percentage change per year with its 95% confidence interval (Slope percent, Lower percent, Upper percent).

Variable	p	Slope	Lower	Upper	Slope percent	Lower percent	Upper percent
Accumulation	0.000	-48.628	-62.282	-34.986	-4.332	-5.198	-3.435
Aitken	0.000	-221.994	-316.781	-131.238	-4.520	-5.674	-2.982
CO	0.003	-0.008	-0.013	-0.003	-3.881	-5.746	-1.603
Mixed traffic	0.000	-101.867	-133.461	-69.754	-4.078	-4.780	-3.175
NO	0.000	-0.133	-0.182	-0.097	-5.192	-6.399	-4.278
NO ₂	0.000	-0.631	-0.784	-0.511	-5.818	-6.693	-5.224
Nucleation	0.003	-135.208	-247.327	-35.142	-3.501	-5.332	-1.060
Nucleation growth	0.003	-29.528	-49.554	-11.345	-3.855	-5.930	-1.717
O ₃	0.284	-0.428	-1.129	0.373	-1.203	-2.949	1.161
Photonucleation	0.083	-64.909	-155.242	6.152	-2.252	-4.469	0.251
PM ₁₀	0.715	-0.077	-0.454	0.311	-0.491	-2.439	2.277
PM _{2.5}	0.240	-0.143	-0.359	0.076	-1.462	-3.380	0.893
PNC	0.000	-397.630	-613.931	-225.436	-4.036	-5.320	-2.626
Regional-1	0.003	-9.998	-19.205	-3.882	-4.018	-6.489	-1.881
Regional-2	0.007	-8.986	-17.034	-3.128	-1.827	-3.141	-0.714
SO ₂	0.003	-0.025	-0.044	-0.007	-3.351	-5.225	-1.157
Total photonucleation	0.013	-108.120	-189.498	-17.763	-2.857	-4.399	-0.553
Total traffic	0.000	-368.969	-464.165	-265.804	-5.009	-5.804	-4.085
Traffic-1	0.000	-140.069	-174.358	-97.133	-5.304	-6.118	-4.071
Traffic-2	0.000	-134.661	-177.651	-101.116	-5.352	-6.645	-4.411

Table S12. Summary of Theil-Sen trend analysis results for the warm season, uncorrected dataset, including particle number concentration (PNC), size modes, source contributions to PNC, and ancillary pollutants. Reported values are the p-value (p), absolute slope (units per year) with its 95% confidence interval (Lower, Upper), and relative slope expressed as percentage change per year with its 95% confidence interval (Slope percent, Lower percent, Upper percent).

Variable	p	Slope	Lower	Upper	Slope percent	Lower percent	Upper percent
Accumulation	0.000	-68.952	-98.240	-48.840	-4.259	-5.566	-3.474
Aitken	0.000	-295.362	-377.044	-205.296	-4.690	-5.675	-3.621
CO	0.000	-0.010	-0.015	-0.008	-3.800	-5.152	-3.078
Mixed traffic	0.000	-136.764	-166.577	-96.171	-4.895	-5.646	-3.892
NO	0.000	-0.260	-0.369	-0.196	-6.584	-7.031	-5.927
NO ₂	0.000	-1.014	-1.211	-0.823	-5.697	-6.503	-5.538
Nucleation	0.003	-135.172	-216.068	-54.838	-3.339	-4.799	-1.562
Nucleation growth	0.013	-26.711	-48.987	-9.413	-3.607	-5.633	-1.503
O ₃	0.708	-0.078	-0.463	0.247	-0.235	-1.327	0.805
Photonucleation	0.033	-77.304	-163.437	-3.431	-2.591	-4.415	-0.138
PM ₁₀	0.751	-0.066	-0.467	0.194	-0.325	-2.084	1.064
PM _{2.5}	0.210	-0.143	-0.420	0.073	-1.091	-2.889	0.633
PNC	0.000	-485.302	-654.413	-337.081	-3.947	-4.904	-3.081
Regional-1	0.007	-8.060	-16.398	-2.339	-4.177	-6.561	-1.461
Regional-2	0.427	-2.567	-9.775	4.845	-0.538	-1.764	1.132
SO ₂	0.000	-0.034	-0.055	-0.017	-4.047	-5.108	-2.255
Total photonucleation	0.007	-118.526	-203.583	-23.737	-3.021	-4.584	-0.734
Total traffic	0.000	-371.978	-465.318	-271.282	-5.076	-5.891	-4.142
Traffic-1	0.000	-132.500	-175.906	-81.743	-5.079	-6.060	-3.682
Traffic-2	0.000	-110.415	-153.882	-81.443	-5.045	-6.410	-4.209

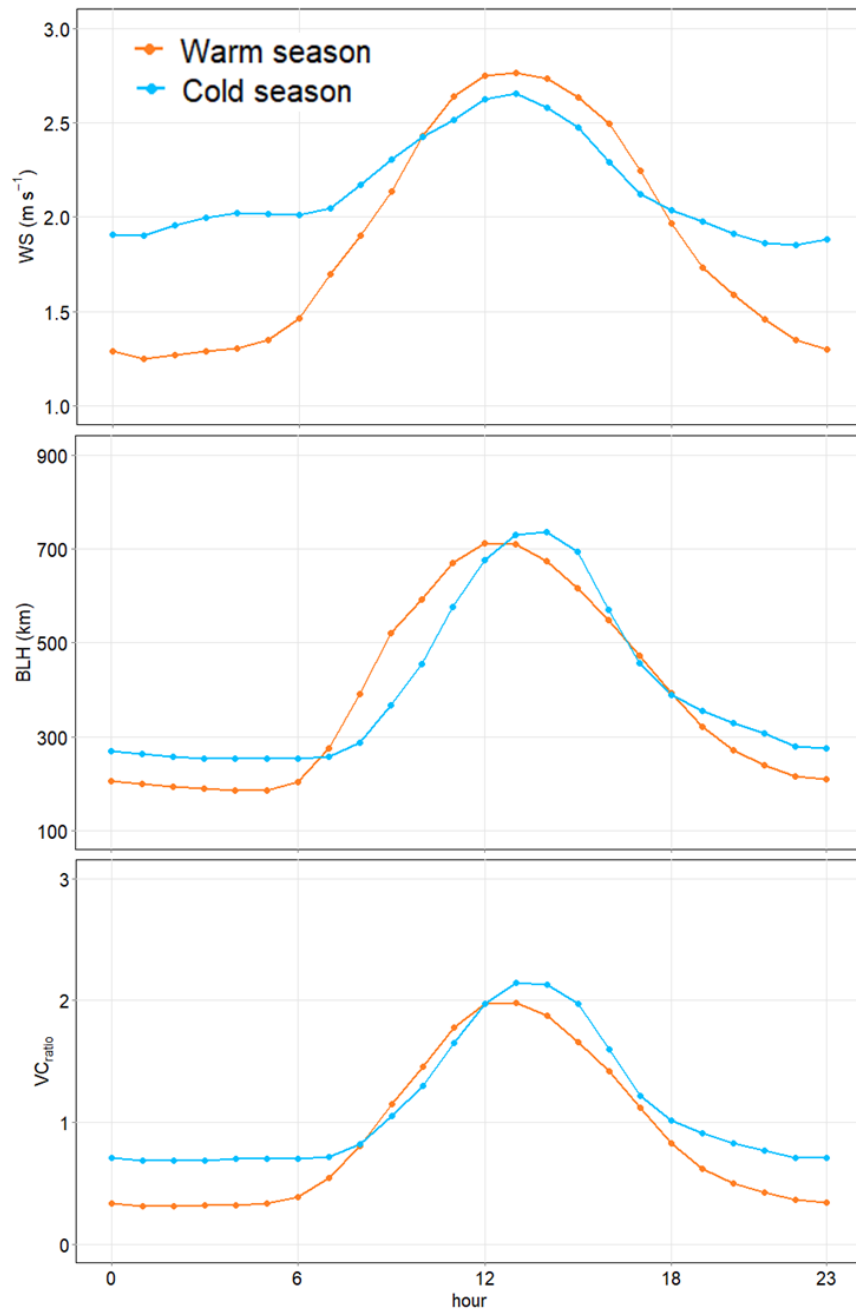


Figure S1. Seasonal mean diurnal variations of (top) wind speed (WS), (middle) boundary layer height (BLH), and (bottom) ventilation coefficient ratio (VC_{ratio}), during warm season (April-September) and cold season (October-March). Values represent hourly means averaged over the full period.

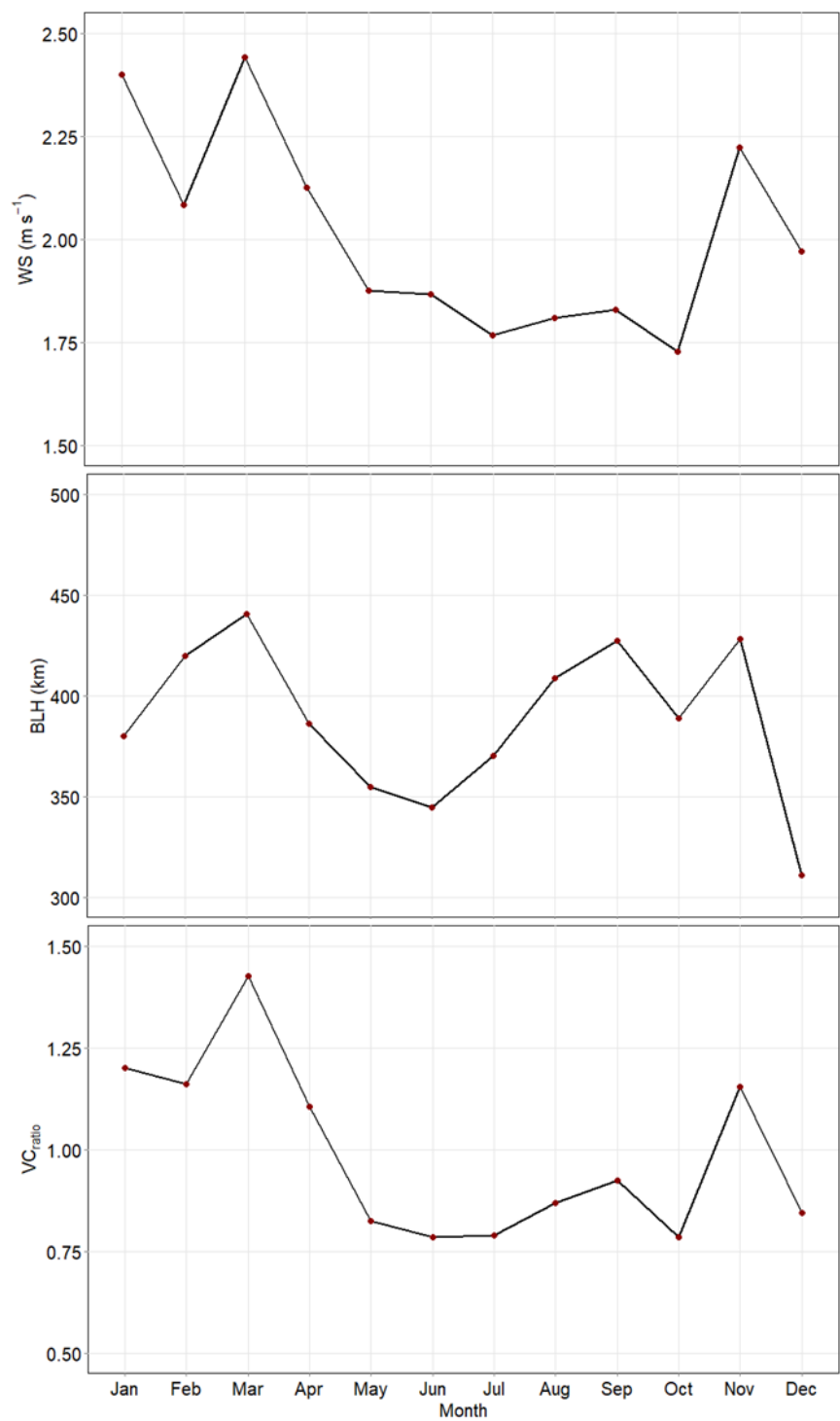


Figure S2. Monthly variations of (top) wind speed (WS), (middle) boundary layer height (BLH), and (bottom) ventilation coefficient ratio (VC_{ratio}). Values represent monthly means averaged over the full period.

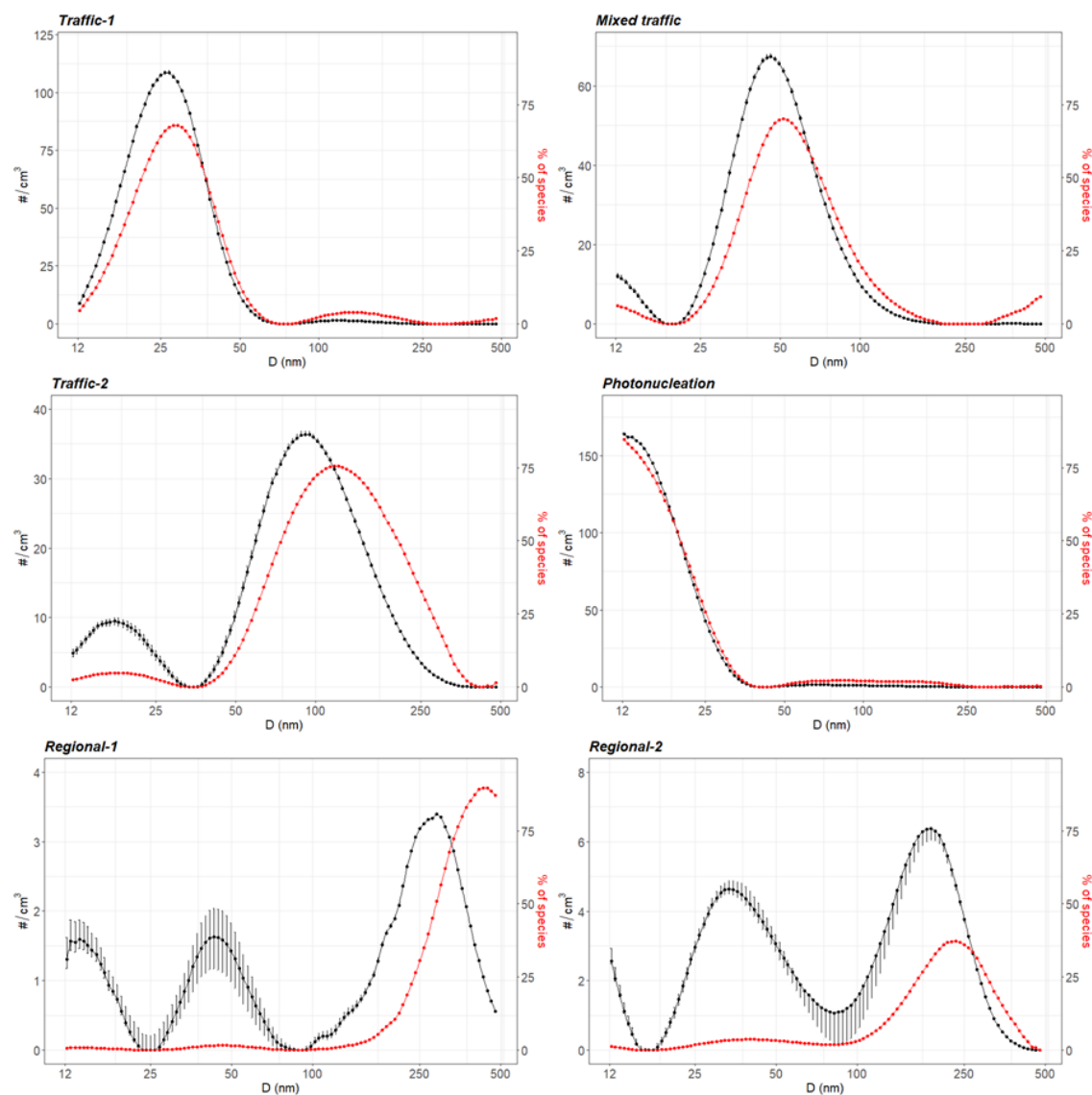


Figure S3. Particle number size distributions (PNSD) of each factor identified by Positive Matrix Factorization (PMF) for the cold season dispersion corrected analysis. The black points represent the particle number concentration (PNC), the error bars are the minimum and maximum displacement (DISP) values, and the red points represent the percentage (%) of explained variation.

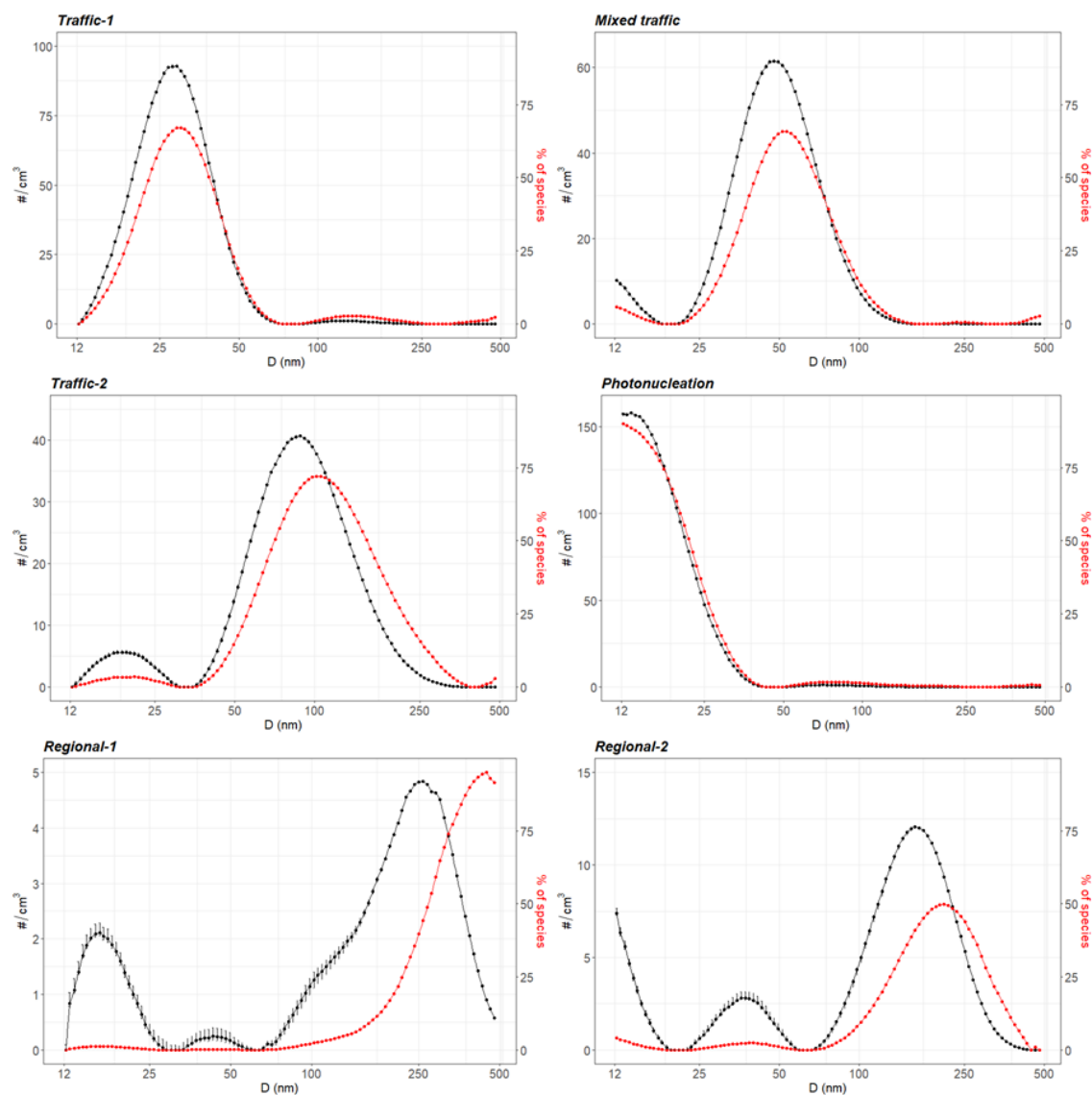


Figure S4. Particle number size distributions (PNSD) of each factor identified by Positive Matrix Factorization (PMF) for the warm season dispersion corrected analysis. The black points represent the particle number concentration (PNC), the error bars are the minimum and maximum displacement (DISP) values, and the red points represent the percentage (%) of explained variation.

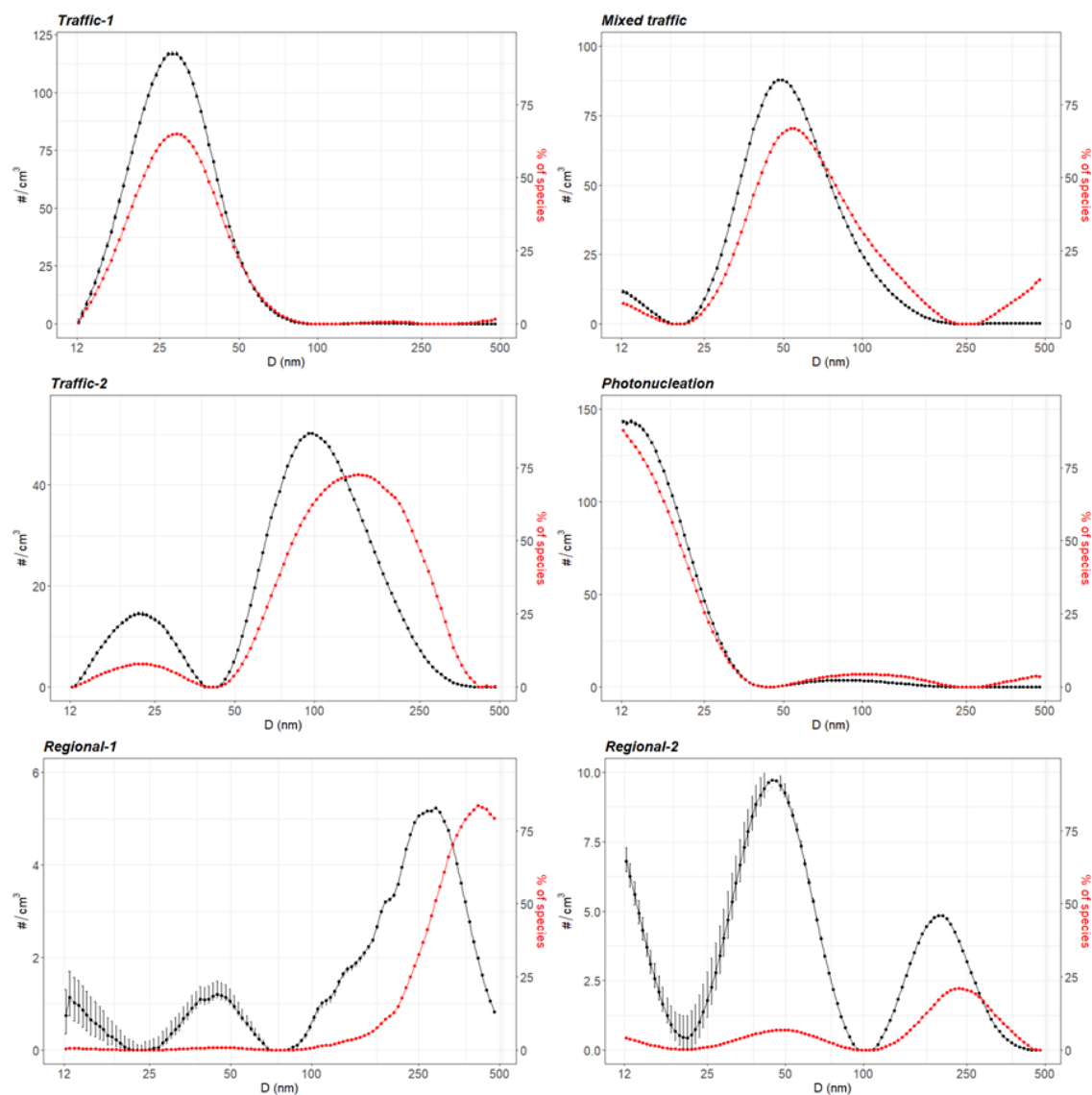


Figure S5. Particle number size distributions (PNSD) of each factor identified by Positive Matrix Factorization (PMF) for the cold season uncorrected analysis. The black points represent the particle number concentration (PNC), the error bars are the minimum and maximum displacement (DISP) values, and the red points represent the percentage (%) of explained variation.

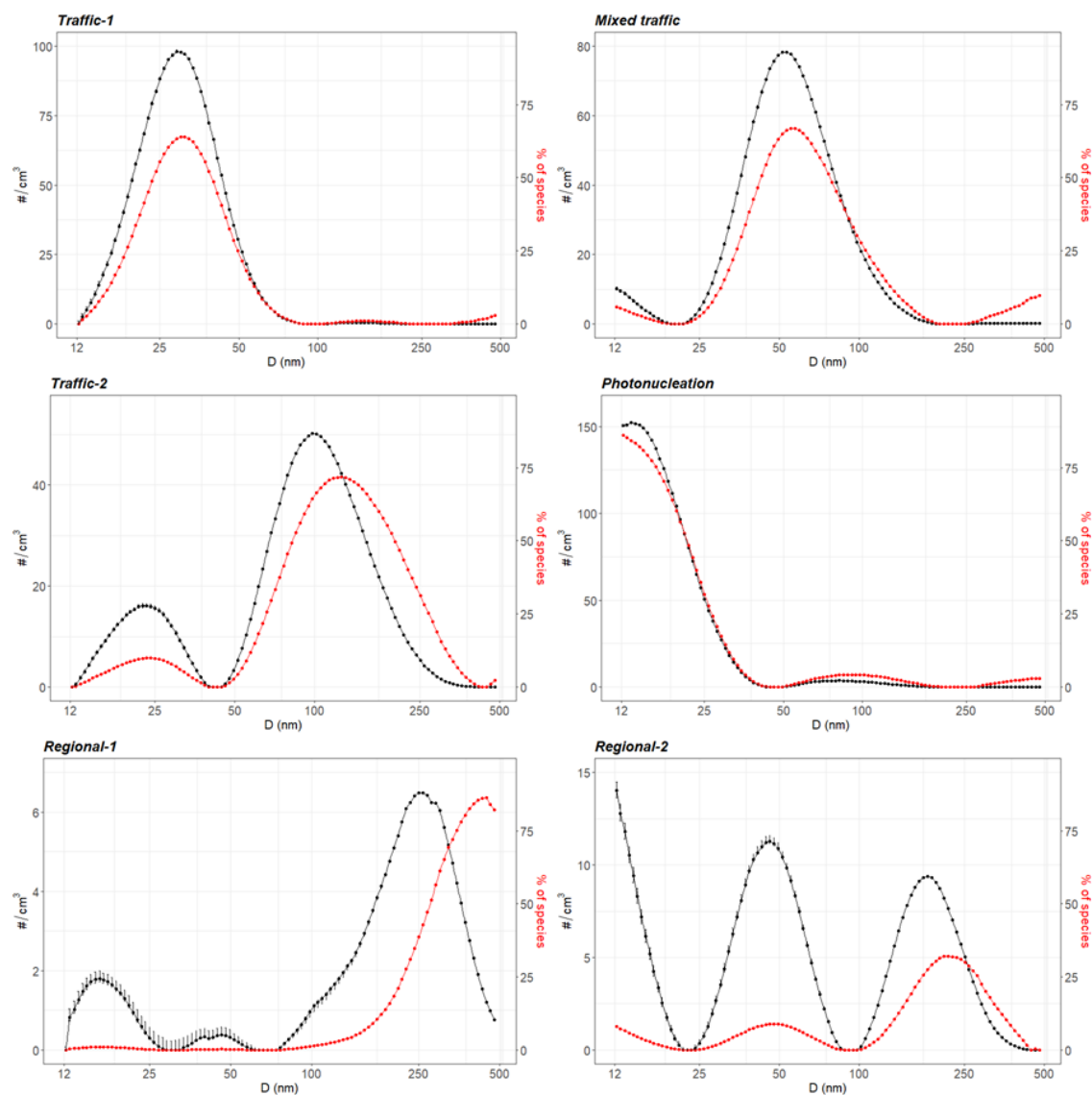


Figure S6. Particle number size distributions (PNSD) of each factor identified by Positive Matrix Factorization (PMF) for the warm season uncorrected analysis. The black points represent the particle number concentration (PNC), the error bars are the minimum and maximum displacement (DISP) values, and the red points represent the percentage (%) of explained variation.

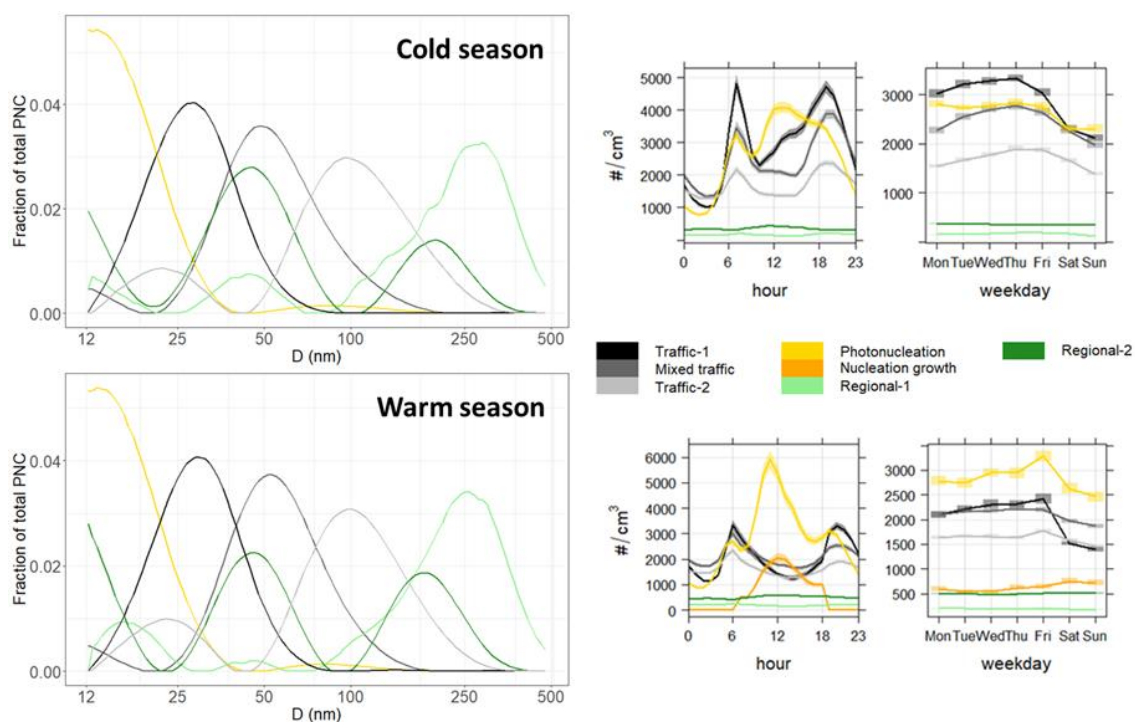


Figure S7. Particle number size distribution (PNSD) profiles (left), and diurnal and weekly patterns (right) of the sources identified in the uncorrected source apportionment for the warm season (April-September, bottom) and the cold season (October-March, top). For the warm season, the profile initially identified as *Traffic-1* (black) was subsequently separated into two distinct sources: *Traffic-1* (black) and *Nucleation growth* (orange).

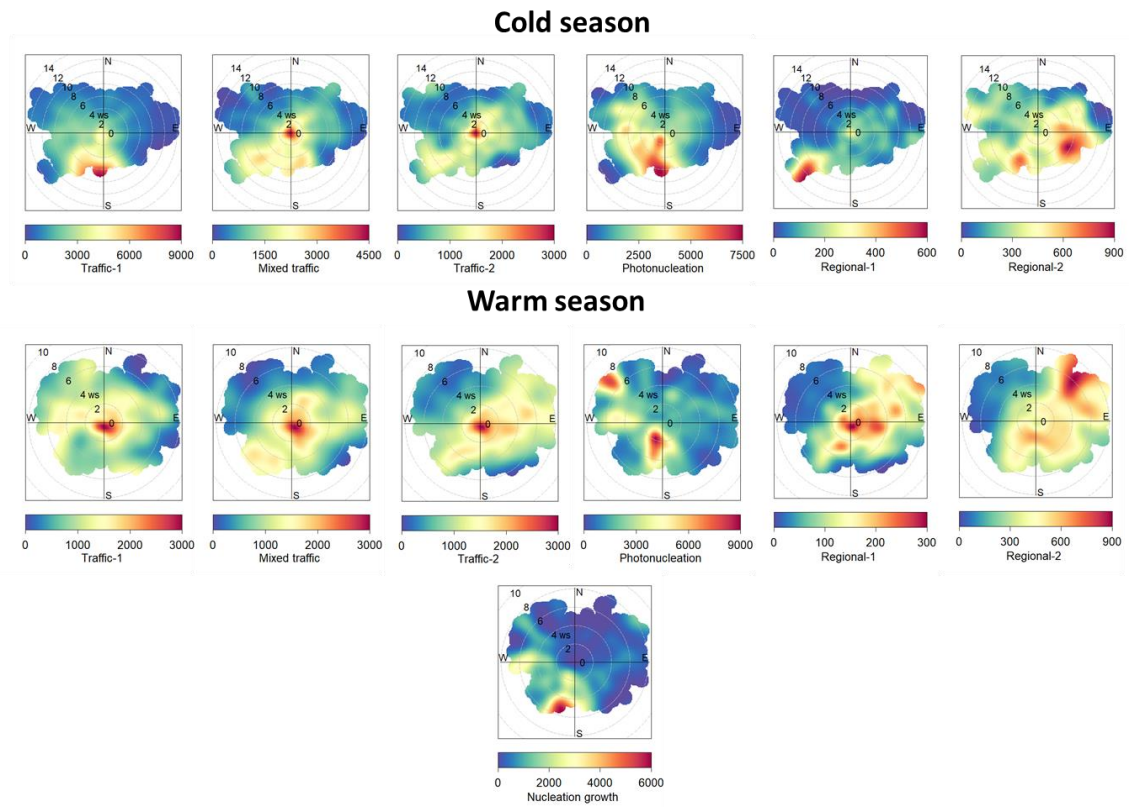


Figure S8. Polar plots showing the source contributions identified in the uncorrected source apportionment for the cold season (October-March, top) and warm season (April-September, bottom).

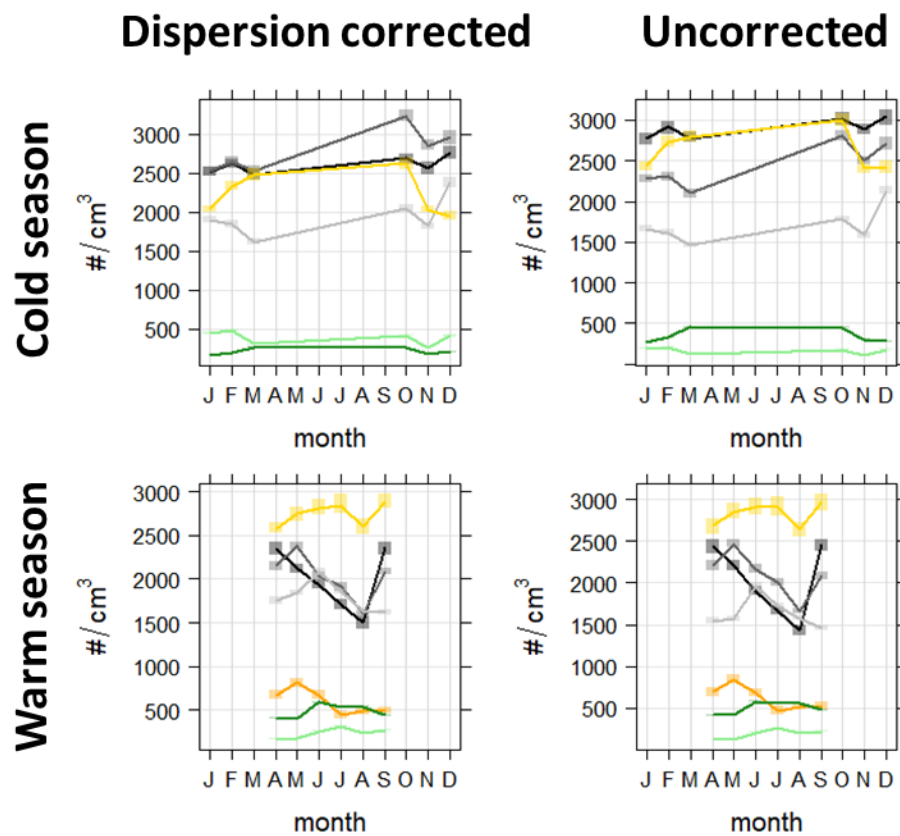


Figure S9. Monthly patterns of the sources identified in the dispersion corrected (left) and uncorrected (right) source apportionment for the warm season (April-September, bottom) and the cold season (October-March, top).

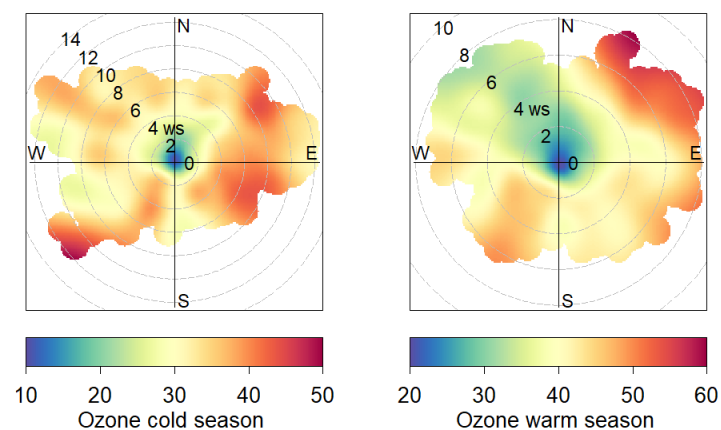


Figure S10. Polar plots showing ozone (O_3) concentrations (ppb) during the cold (October-March, left) and warm season (April-September, right).

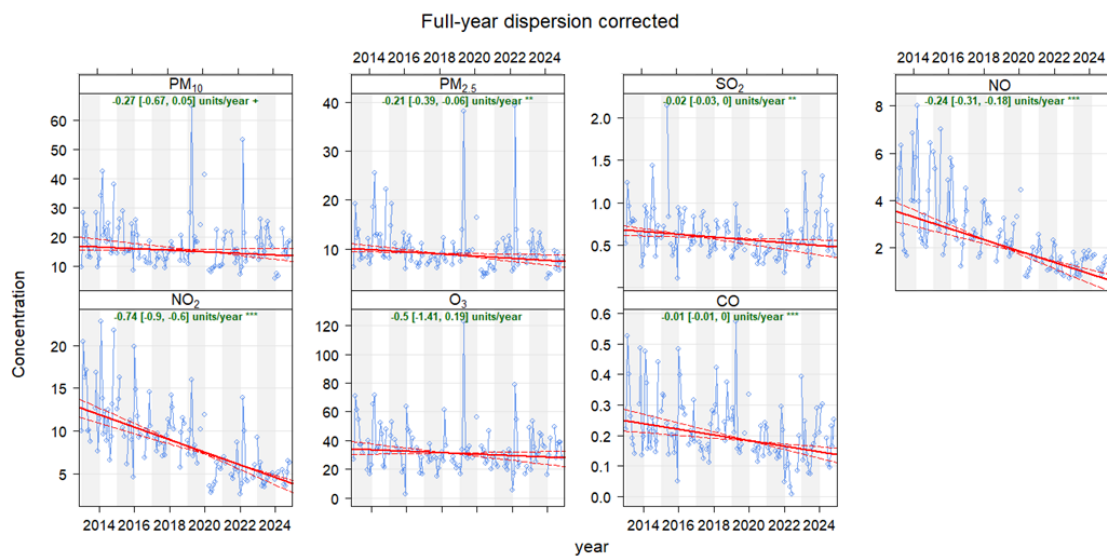


Figure S11. Trend analysis of full-year, dispersion corrected ancillary pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$), and “+” ($p < 0.1$).

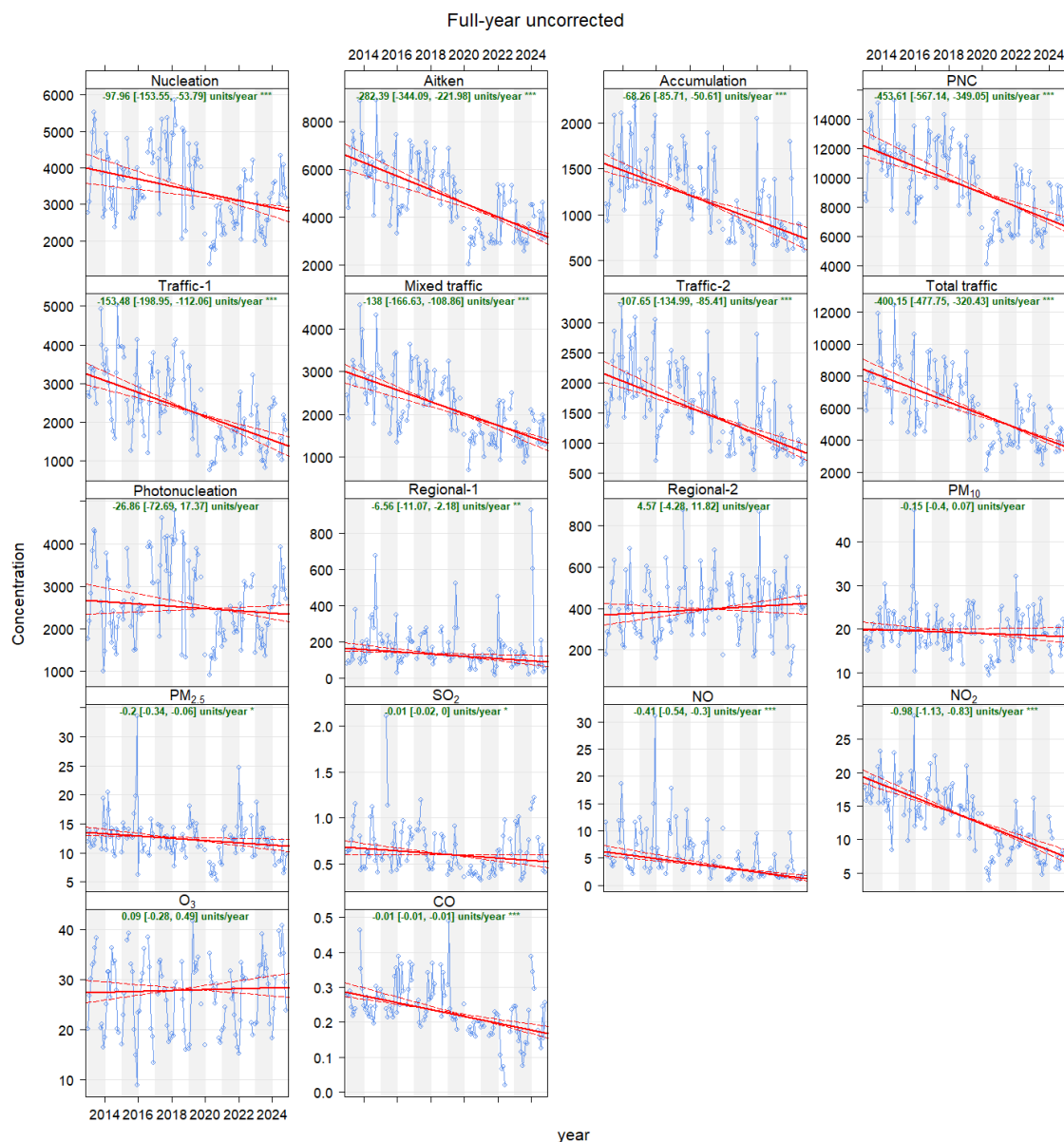


Figure S12. Trend analysis of full-year, uncorrected particle number concentration (PNC) and size modes (Nucleation, Aitken, and Accumulation), source contributions to PNC (Traffic-1, Mixed traffic, Traffic-2, Total traffic, Photonucleation, Nucleation growth, Regional-1, and Regional-2), and ancillary pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “****” (p<0.001), “***” (p<0.01), “**” (p<0.05), and “+” (p<0.1).

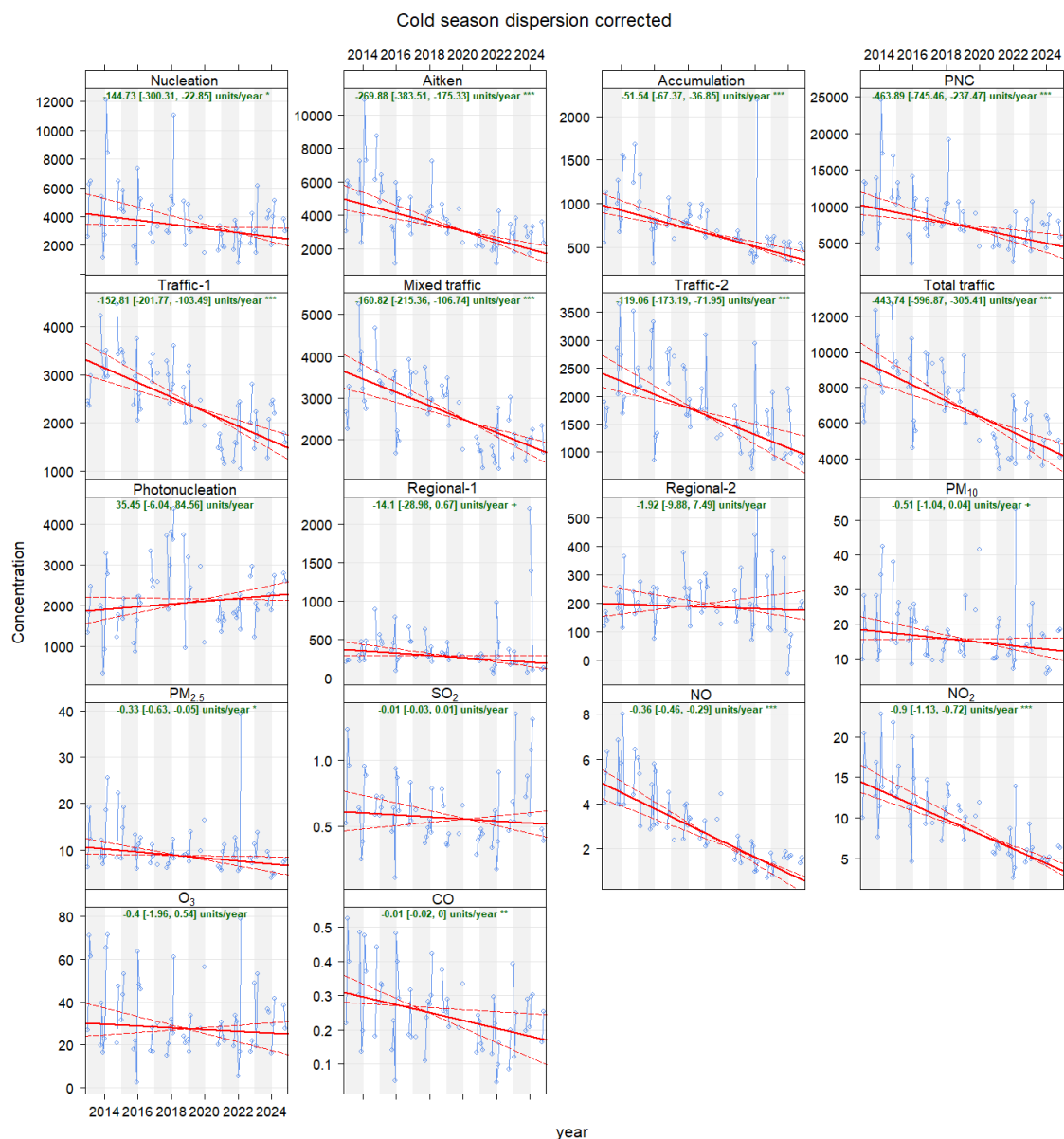


Figure S13. Trend analysis of cold season, dispersion corrected particle number concentration (PNC) and size modes (Nucleation, Aitken, and Accumulation), source contributions to PNC (Traffic-1, Mixed traffic, Traffic-2, Total traffic, Photonucleation, Regional-1, and Regional-2), and ancillary pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “****” (p<0.001), “***” (p<0.01), “**” (p<0.05), and “+” (p<0.1).

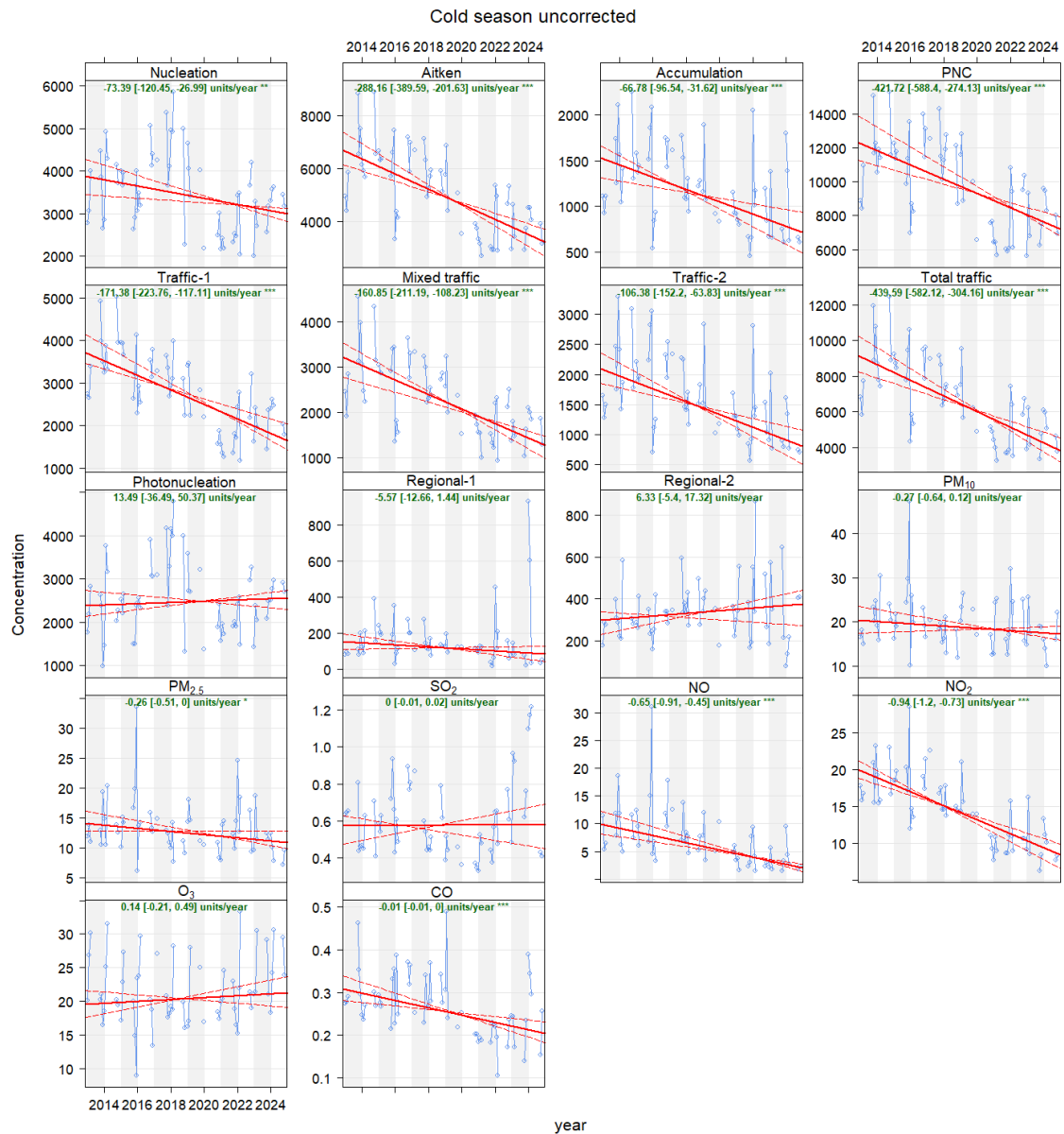


Figure S14. Trend analysis of cold season, uncorrected particle number concentration (PNC) and size modes (Nucleation, Aitken, and Accumulation), source contributions to PNC (Traffic-1, Mixed traffic, Traffic-2, Total traffic, Photonucleation, Regional-1, and Regional-2), and ancillary pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “***” (p<0.001), “**” (p<0.01), “*” (p<0.05), and “+” (p<0.1).

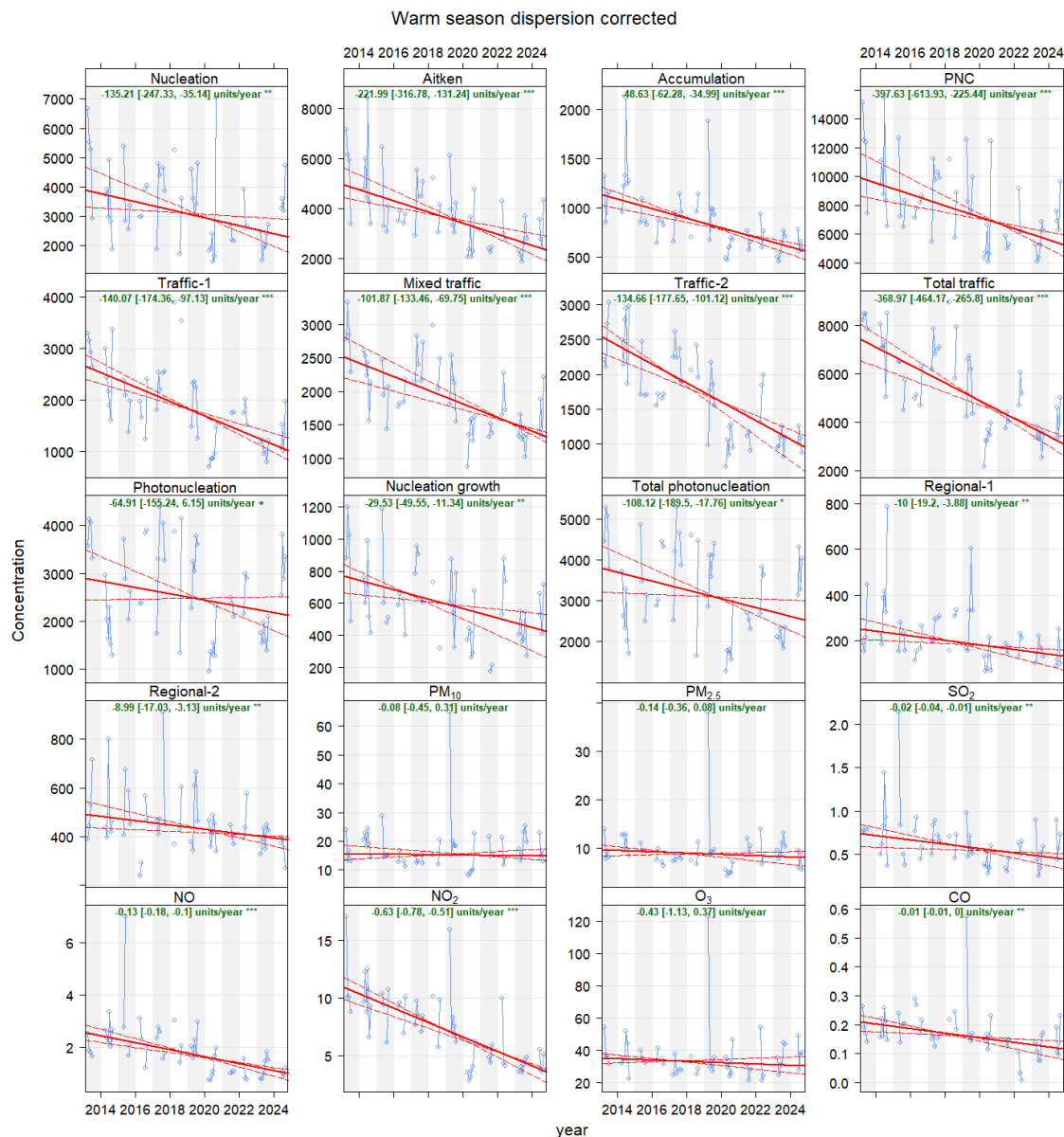


Figure S15. Trend analysis of warm season, dispersion corrected particle number concentration (PNC) and size modes (Nucleation, Aitken, and Accumulation), source contributions to PNC (Traffic-1, Mixed traffic, Traffic-2, Total traffic, Photonucleation, Nucleation growth, Total photonucleation, Regional-1, and Regional-2), and ancillary pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “***” (p<0.001), “**” (p<0.01), “*” (p<0.05), and “+” (p<0.1).

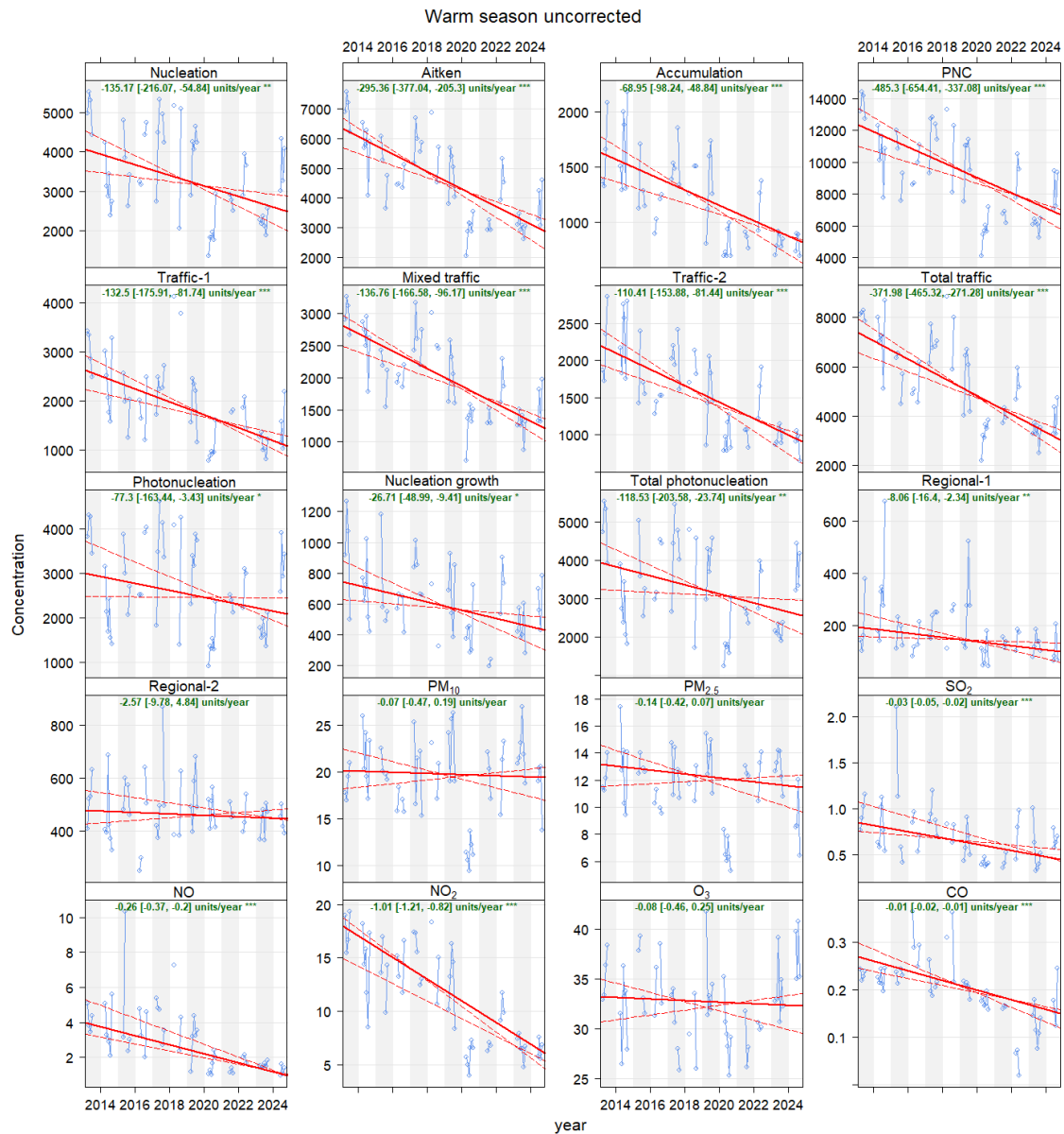


Figure S16. Trend analysis of warm season, uncorrected particle number concentration (PNC) and size modes (Nucleation, Aitken, and Accumulation), source contributions to PNC (Traffic-1, Mixed traffic, Traffic-2, Total traffic, Photonucleation, Nucleation growth, Total photonucleation, Regional-1, and Regional-2), and ancillary pollutants (PM₁₀, PM_{2.5}, SO₂, NO, NO₂, O₃, and CO). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “***” (p<0.001), “**” (p<0.01), “*” (p<0.05), and “+” (p<0.1).

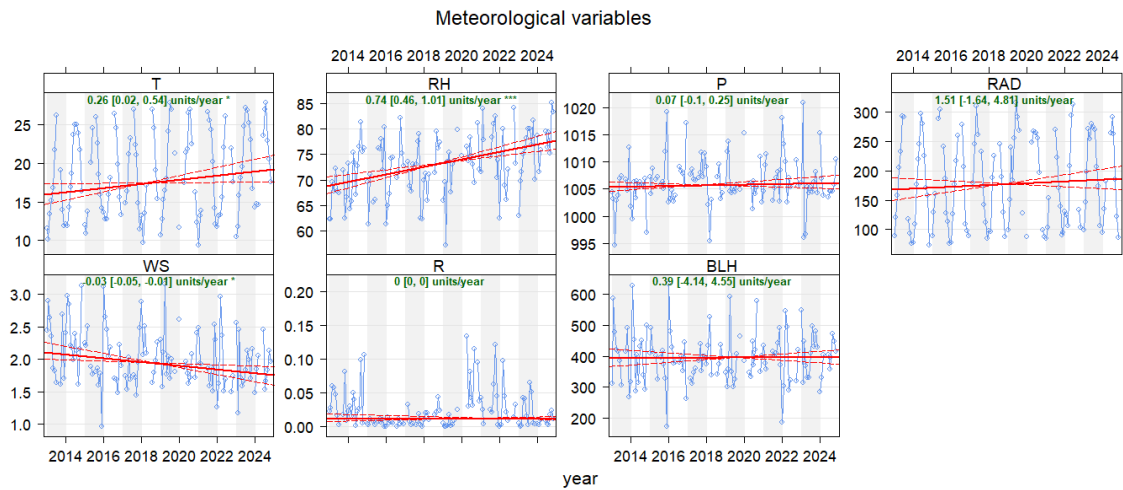


Figure S17. Trend analysis of meteorological variables (T, temperature, in °C; RH, relative humidity, in %; P, pressure, in hPa; WS, wind speed, in m/s; R, rain, in mm; BLH, boundary layer height, in m). Theil-Sen trend slopes with 95% confidence interval are shown on the plot. Statistical significance is indicated by “***” ($p < 0.001$), “**” ($p < 0.01$), “*” ($p < 0.05$), and “+” ($p < 0.1$).

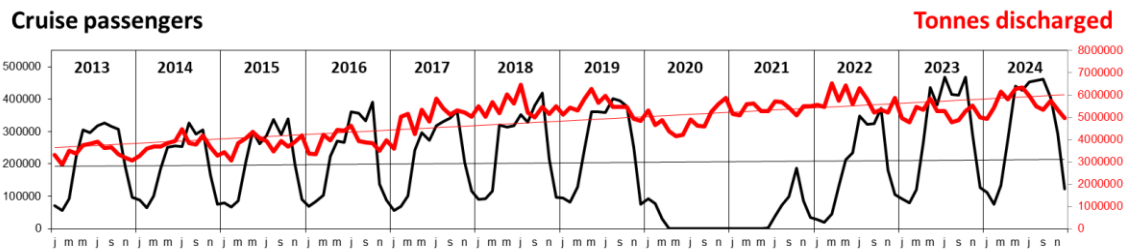


Figure S18. Monthly cruise passenger numbers and tonnes of discharge in the harbour of Barcelona, according to Port of Barcelona (2025).

References

Port de Barcelona, 2025. Estadísticas del Puerto de Barcelona. <https://www.portdebarcelona.cat/es/web/autoritat-portuaria/estadistica>



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