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Measurements of Particulate Matter from Electronic and Conventional Cigarettes: A Comparative Analysis of Methods

Ario Ruprecht [1,2], Alessandro Borgini [2,3,*], Chiara Veronese [1], Andrea Tittarelli [4], Roberto Boffi [1], Martina Bertoldi [3], Esteve Fernández [5,6,7,8], Cinzia De Marco [9] and TackSHS Project Investigators [†]

1 Respiratory Disease Unit, Fondazione IRCCS Istituto Nazionale Tumori, 20133 Milan, Italy
2 International Society of Doctors for Environment (ISDE), 52100 Arezzo, Italy
3 Environmental Epidemiology Unit, Fondazione IRCCS Istituto Nazionale dei Tumori, 20133 Milan, Italy
4 Cancer Registry Unit, Fondazione IRCCS Istituto Nazionale dei Tumori, 20133 Milan, Italy
5 Tobacco Control Unit, Institut Català d'Oncologia, Av. Granvia de l'Hospitalet 199-203, 08908 L'Hospitalet de Lobregat, Spain
6 Tobacco Control Research Group, Institut d'Investigació Biomèdica de Bellvitge, Av. Granvia de l'Hospitalet 199, 08908 L'Hospitalet de Llobregat, Spain
7 School of Medicine and Health Sciences, Universitat de Barcelona, Feixa Llarga, s/n, 08907 L'Hospitalet de Llobregat, Spain
8 Center for Biomedical Research in Respiratory Diseases (CIBERES), Av. Monforte de Lemos, 3-5, Pabellón 11, Planta 0, 282029 Madrid, Spain
9 Biomarkers Unit, Fondazione IRCCS Istituto Nazionale dei Tumori, 20133 Milan, Italy
* Correspondence: alessandro.borgini@istitutotumori.mi.it; Tel.: +39-0223903536
† Membership of the TackSHS Project Investigators is provided in the Acknowledgments.

Abstract: Due to the growing popularity of electronic cigarettes (ECs) and heated tobacco products (HTPs) as alternatives to conventional cigarettes (CCs), there is an increasing need to monitor the emissions of these new devices. ECs generate significant concentrations of second-hand aerosol (ECSHA), which is visible in dense clouds and can be smelled. Particulate matter (PM) is an important component of CC, HTP and EC aerosols, and Optical Particle Counters (OPCs) enable its real-time measurement, which is expressed either as the number of particles or as mass. This study specifically addresses the limitations associated with EC mass measurement using OPC technology and identifies the strict necessity of the measurement of a corresponding density (k factor) not only for each specific PM source but also for the desired PM size. Therefore, a standard measurement requires the simultaneous operation of the OPC equipment and a certified reference instrument. Four different OPCs were used. Crucially, this study also proves that this setup may be inapplicable because the extreme volatility of EC-generated aerosols makes it impractical to gauge the correct EC k factor.

Keywords: electronic cigarettes; second-hand aerosol; $PM_{2.5}$; particle counter; calibration factor

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1. Introduction

The use of electronic cigarettes (ECs) and heated tobacco products (HTPs) [1,2] generates significant concentrations of second-hand aerosol (SHA) [3–6] that may sometimes be visible in very dense clouds and may also be smelled. It is well known that the use of ECs significantly contributes to the environmental deterioration of air quality, particularly indoors [7–16]. Numerous studies have shown significant harmful health effects in EC users [17–20] in addition to negative health consequences in those exposed to ECSHA, particularly in sensitive subjects such as children [21], pregnant women and the elderly. Real-time Optical Particle Counter (OPC) measurements, expressed as mass concentration ($\mu g/m^3$) of PM, are frequently used [22,23], particularly to measure second-hand smoke (SHS) from conventional cigarettes (CCs). As OPCs can also produce measurements expressed as particle number (counts/cm^3 or counts/L) [24], in this study, funded by the TackSHS research project and part of the European Union's Horizon 2020 program [25], we investigated the pros and cons associated with these two different OPC outputs.

1.1. OPC Principle of Operation

OPC instruments are generally equipped with a system consisting of a sampling pump that draws the air to be measured into a system composed of a chamber containing a laser source, lenses, mirrors, detector and electronic parts. The purpose is to convert the laser radiation reflected by all particles present in the sampled air and passing through the laser beam into electric signals. These signals are classified into different levels and durations corresponding to different particle size ranges, regardless of their physical status as a liquid, semiliquid or solid. Almost all OPC models are provided with a programmable sampling time from a few seconds to minutes or hours and with internal memory. The exhaust sample from the pump, rotary vane with graphite blades or membrane models is filtered to avoid environmental contamination from these pump components and possible recirculation to the inlet. However, low-cost OPCs use fans without exhaust filtration. Many OPCs are also equipped with sample heaters to automatically control the relative humidity (RH) in the laser chamber [26,27].

1.2. OPC Calibration for Output as Number of Particles

The manufacturer calibration is performed on a number of particles for each programmed size according to accepted industry methods using equipment, procedures and standards that are traceable to the National Institute of Standard and Technology (NIST) and the International Organization for Standardization (ISO). Usually, polystyrene latex particles are used to simulate different PM pollution levels and size profiles [27,28]. For instance, for model 212-2's factory calibration, the polystyrene latex particle (PSL) sizes are (μm): 0.5, 0.7, 1.0, 2.0, 3.0, 5.0, 7.0 and 10.0, with accuracy +/− 10% and reporting the Lot# NIST [29].

1.3. OPC Calibration for Output in Mass

To convert the number of particles into mass, it is necessary to know the PM density. The particle density can be considered an empirical function of the characteristics of the light reflected onto the detector by each particle as the pulse intensity and duration. This density k factor is usually set by the manufacturer at 1.000 and refers to the polystyrene latex particle density [30]. However, environmental PM physical and chemical composition is different and extremely variable and strongly correlates with emission sources, weather conditions (temperature, relative humidity, wind speed and solar radiation) and other factors [31–35]. These physical and chemical changes influence not only the density but also the optical properties of the PM, and consequently, it becomes necessary to apply a different density k factor to the equation for each type of PM. For example, the k factor applied to measure summer urban PM pollution, predominantly generated by traffic, will not be correct for PM measurements performed in winter because of the building heating contribution to urban pollution, which significantly changes the PM chemical composition and consequently its optical properties. Therefore, it is imperative to adequately correct for each specific condition of measurement following a procedure that inherently takes into account the conditions of operation of the OPC in addition to a reference method.

Besides the above-mentioned k factor variability, additional variation may also be introduced by the use of different instruments, even if they are the same model and from the same manufacturer. This discrepancy may be due to individual differences in laser wavelength, intensity, optical properties of lenses, mirrors, detectors, etc., which may cause a different response of the light diffracted by the particles. Consequently, each instrument must be individually calibrated to find its own individual k factor [36]. For the reasons explained, a comparison of results across different studies may be flawed, even if performed with the same OPC model, when the instrument is used by simply applying the k factor set by the manufacturer.

Several studies have been conducted that measured PM using OPC-detected mass [7,12,37–41]. Most of them used the model TSI SidePak. However, the calibration of

the OPC is not mentioned, and therefore, we suppose that the factory density k factor was used with all of the drawbacks described in our study.

Environmental RH [42,43] is an important interference factor in OPC measurement because it introduces significant and non-linear changes in the particle volume and optical properties and must be taken into consideration and compensated (see Figures S1 and S2 in Supplementary Information).

2. Materials and Methods

2.1. Sampling Site

All measurements were performed at the Fondazione IRCCS Istituto Nazionale dei Tumori, Milan, Italy (INT), laboratory, in a 48 m^3 room with a measured Air Exchange Rate (AER) of 0.7/0.8 per hour, with the instruments installed on a table about 2 m away from the volunteer smoker or user of the CC or EC. The laboratory is furnished with typical home appliances (e.g., closets, tables, and chairs) and is equipped with one individual (not shared with any other offices or laboratories nearby) Air Conditioning System (ACS) characterized by indoor air recirculation of about 500 m^3/h without filters. During the experiments, the room was occupied by two or three persons, one of whom was either using the CC or EC. One mechanical fan was always in operation to create air turbulence in order to ensure that the aerosol and PM concentrations were, as much as possible, the same in every part of the room, reaching the maximum mixing factor on a scale from 0 to 1. Temperature (T) and relative humidity (RH) were continuously measured during all tests. The instruments were installed on a table on one side of the room, and the vapers/smokers were generally seated on the other side at about three meters away or walking up and down to simulate the real-world behavior of people in a closed environment.

2.2. Evaluation of OPCs to Measure EC Second-Hand Aerosol (ECSHA) Mass

2.2.1. Instruments

Reference instrument: Model BAM-1020 manufactured by Metone Instruments Inc. (Equivalent Method Application No. 07-262-11; Met One BAM-1020 Monitor—$PM_{2.5}$/PM_{10} FEM Configuration and TÜV RHEINLAND IMMISSIONSSHUTZ UND ENERGIESYSTEME GMBH).

OPC instruments used for mass measurements in μg/m^3 (all previously calibrated for the density correction factor of SHS from CC with the same BAM-1020) are presented in Table 1 [44,45].

Table 1. Details of the OPC analyzers used and the reference tool, BAM-1020.

Model	Technical Features	Measurements
Dylos DC 1700	OPC (Optical Particle Counter)	Particles/ft^3 of sizes between >0.5 and >2.5 μm
Air Visual	OPC (Optical Particle Counter)	PM_1, $PM_{2.5}$, PM_{10}, temperature and relative humidity
AirBeam	OPC (Optical Particle Counter)	PM_1, $PM_{2.5}$, PM_{10}, temperature and relative humidity
Aerocet 531S	Full-featured OPC (Optical Particle Counter)	PM_1, $PM_{2.5}$, $PM_{4.0}$, $PM_{7.0}$, PM_{10} and TSP
BAM-1020	Beta attenuation mass monitor	$PM_{2.5}$ and PM_{10}

2.2.2. Calibration Procedure

OPC mass calibration can be performed using the Beta Attenuation Monitor (BAM) as a reference method, as performed in this study, or equivalent. The BAM principle of measurement is that the attenuation of electrons emitted by a radioactive source (usually ^{14}C) by PM accumulated on a filter tape is sensitive to the H_2O present in the PM, and consequently, the sample must mandatorily be equipped with an automatic heater. The RH % must be set to 35% for the version of the BAM employing a smart heater when operated as a $PM_{2.5}$ US-EPA-designated federal equivalent method. The RH set point is set to 45% for European (EU) $PM_{2.5}$ units. The following are two different OPC mass calibration procedures [46].

(a) If the OPC is equipped with sample automatic heat and RH % control, it should be set at the same RH % as the BAM for accurate calibration. However, in the case of the presence of volatile or semi-volatile elements in the PM, there may be errors in the k factors because they evaporate on both instruments.

(b) If the OPC is not equipped with heaters or RH sensors, calibration with the BAM must be performed by measuring the environmental RH with an adequate RH sensor and applying mathematical Equation (1), an empirical adjustment equation derived from the experimental data of Laulainen [47], which fits well with data from several studies [48], and then compared with the reference instrument measurements. However, it must be considered that the RH interference becomes extremely high when it increases above 80/85% (see Figures S1 and S2 and Table S1 in SI—Supplementary Information), causing unacceptable errors in the mathematical interference correction, mainly because of uncertainties in the measurements at these high RH levels. If the OPC must be used when the RH is above 80/85%, sample dryers or heaters become compulsory.

$$PMc = PMm \times a + \frac{b \times RH^2}{1 - RH} \quad (1)$$

where

PMc = PM compensated for RH;
PMm = OPC PM hourly average measured values;
a = 1.0, as reported by Chakrabarti et al. [48];
b = 0.25, as reported by Chakrabarti et al. [48];
RH = RH in the 0–0.99 range, corresponding to 0–99% or RH/100.

The density k factor is calculated by applying Equation (2).

$$k = PMr/PMm \quad (2)$$

where

k = density k factor;
PMr = BAM-1020 PM hourly average reference values;
PMm = OPC PM hourly average measured values.

However, in the case of the presence of volatile or semi-volatile elements in the PM, there may be errors in the k factors because they evaporate on the BAM but not on OPCs.

The k factors suitable for measurements of the indoor pollution generated by the different CCs or HTP cigarettes are experimentally found following standard procedures, which mainly consist of operating the OPC to be calibrated in parallel with one density reference instrument in an indoor environment. Two reference methods may be used to find the appropriate density k factors for the OPC:

(a) Comparison of OPC time-integrated mass measurements with the mass of PM accumulated on a pre-weighed filter, following the procedures according to BS EN 12341:2014.

(b) OPC real-time measurement comparison with quasi-real-time measurements by an automatic density equivalent method.

In this study, we followed the second procedure (b) using the model BAM-1020 with US EPA and T.Ü.V. certificates (see specifications in SI) based on the Beta Attenuation Principle. The reason for this choice is that this method presents the advantage of performing mass measurements with a sampling time of one hour and within a wide range of concentration values, while the classical procedure in BS EN 12341:2014 provides two points only: intercept and slope. This procedure allows the evaluation of not only the intercept and slope of the OPC measurements but also the linearity response, precision and accuracy by applying a mathematical regression equation analysis.

One volunteer habitual smoker or user of e-cig started to smoke or to use the EC ad libitum for three hours at the described site. Then, the volunteer left the room to allow the PM concentrations to slowly decrease to the background level due to the room's natural AER, gravitational sedimentation and wall absorbance. After PM concentration stabilization, the OPC measurements were corrected for RH interference according to Equations (1) and (2).

2.2.3. Evaluation of Gravimetric Calibration of OPCs

During all tests, the environmental RH never exceeded 45/50%, and consequently, the OPC measurements were only moderately affected by RH interference, but sample heating on the BAM-1020, according to the certified procedures, was always in operation to keep the sample RH < 25%.

Basic Dylos measurements are performed based on particles/ft^3 for sizes between >0.5 and >2.5 μm. The conversion of particle number concentration (PNC) to $PM_{2.5}$ mass concentration was calculated by subtracting PNC > 2.5 μm from PNC > 0.5 μm and applying the density k factor equation produced by regression according to the Semple equation [41]:

$$\text{Dylos } PM_{2.5} = (ax^2 + bx + c) \times k \quad (3)$$

where:

$PM_{2.5}$ is the mass concentration in $\mu g/m^3$;
x is PNC > 0.5 minus PNC > 2.5 μm per ft^3 measured by the Dylos device;
a, b, c and k are coefficients to be determined after calibration in CC SHS with the BAM-1020.

The Semple equation is based on over 500,000 min of data from 34 smoking homes where a Sidepak was co-located next to a Dylos instrument. The resulting best-fit regression equation enables the conversion of the Dylos particle number concentrations to a $PM_{2.5}$ mass equivalent, as measured by the Sidepak device. The homes used in the study are broadly representative of typical smoking homes in northern Europe and represent a range of concentrations, temperatures and humidity levels throughout the measurements.

This procedure has also been proven successful in other studies [49] and produces excellent and repeatable results in the calibrations of all OPC models when used to assess the extent of SHS from CC mass PM exposure. All other OPC models (Air Visual for $PM_{2.5}$, Airbeam for $PM_{2.5}$, and Aerocet 531 for PM_1, $PM_{2.5}$, $PM_{7.0}$, PM_{10} and TSP) employed in this study were calibrated in SHS from CC following the same procedure, and for each instrument, the proper density k factor was determined and applied in all other tests.

Three ECs brands were used: Elips C, Just Fog and JUUL. Several volunteers, all regular users of e-cig, used the ECs "ad libitum" in order to simulate a real-life situation as much as possible.

2.3. Evaluation of OPCs to Measure ECSHA in Number of Particles

In this study, we used one OPC and one CPC model with the concentration expressed in particles/cm^3 and particles/L. Of course, this solution presents the errors indicated by the manufacturer, +/− 20% and +/− 10% for the TSI 3007 and the 212-2 Profiler, respectively. In this study, we included the environmental RH in all measurements, but it never exceeded 45/50%; therefore, we did not apply any RH interference corrections, and the heater of the Profiler 212-2 instrument was switched off.

Instruments

The CPC and OPC instruments used for measurements in number of particles (manufacturer factory-calibrated) were:

- Portable CPC model TSI 3007 measuring PM in counts/cm^3 from 10 to 1000 nanometers; concentration accuracy +/− 20%;
- Bench-top Metone Instruments Inc. model 212-2 Profiler (OPC) measuring PM in counts/L in 8 channels programmed as: >0.3, >0.5, >0.7, >1.0, >2.5, >3.0, >5.0 and

>10.0 µm; programmable sampling time and equipped with heater for automatic RH % control; and concentration accuracy +/− 10%.

2.4. Statistical Procedures

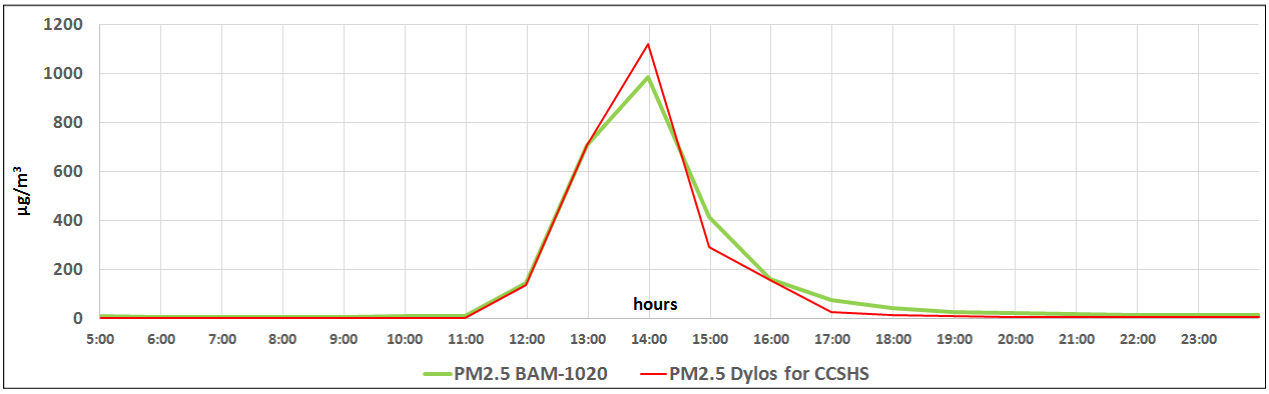
For each calibration session, we calculated the background $PM_{2.5}$ mean and standard deviation (SD) before, during and after CC smoking and e-cig use; we also calculated the Pearson R correlation between the two instruments' values (reference BAM and each OPC to calibrate). As the $PM_{2.5}$ measurements were not normally distributed, we used a non-parametric test, such as the Wilcoxon Signed-Rank test, to evaluate the differences between the two instruments to compare them (null hypothesis to be tested—H_0: the means are equal).

The handling of data and statistical analyses were performed using Microsoft Excel (version 2007) and SPSS (version 17.0), respectively.

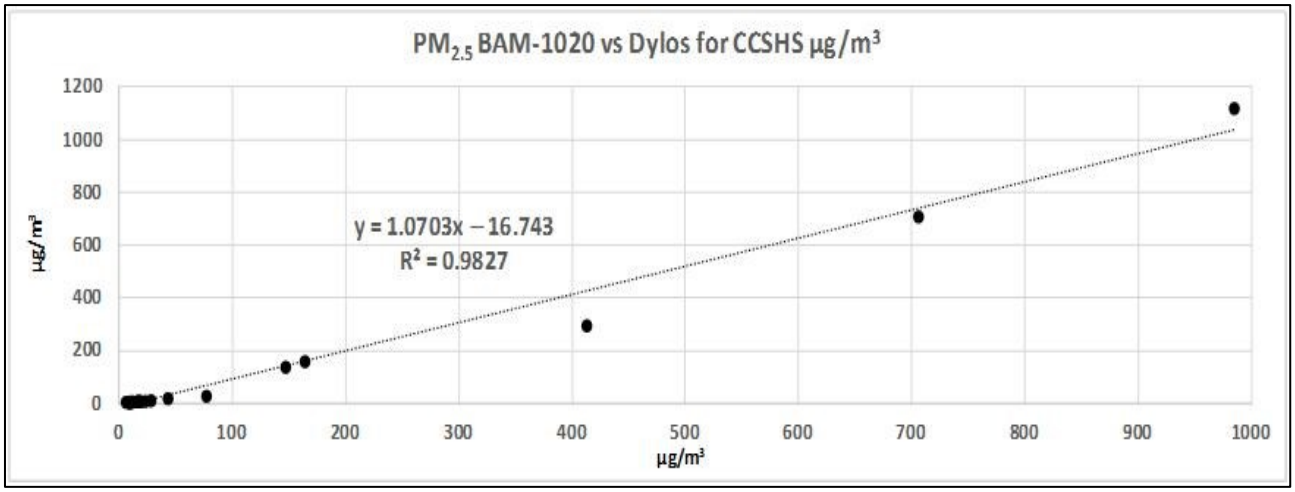
3. Results

3.1. Measurements in Mass

In Figure 1 and Table S1, an example of the results of the calibration (in mass of $PM_{2.5}$) of one OPC (the model Dylos 1700 S/N 351) in SHS from the CC environment is shown. Equation (3) was applied using a value of k = 2.48.



(**a**)



(**b**)

Figure 1. Results of the calibration (in mass of $PM_{2.5}$) of OPC in SHS from CC: real-time graph (**a**) and linear regression analysis (**b**).

Figure 2 presents an example of the results of the same OPC (the model Dylos 1700 S/N 351) calibrated in mass of SHS from CC $PM_{2.5}$ but operating in an ECSHA environment

generated by the EC brand Elips C. Pearson R was 0.465, while the Wilcoxon Signed-Rank test *p* value was 0.374.

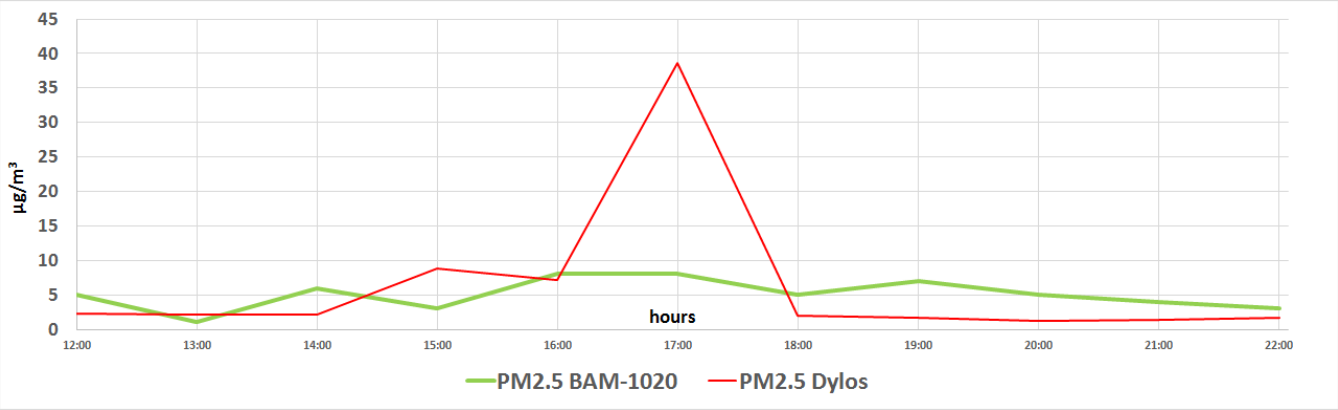


Figure 2. Real-time graph of OPC operating in an ECSHA environment generated by the EC (brand Elips C).

No density k factor and regression analysis were possible because the BAM-1020 did not record any appreciable increment attributable with certainty to ECSHA during the use of the e-cig session.

The BAM-1020's average $PM_{2.5}$ during the e-cig session showed only a minor increase from the background, from 4.0 to 6.3 µg/m^3, while the Dylos measurements increased from 2.2 to 18.3 µg/m^3, with a maximum value of 38.7 µg/m^3 (see Table 2).

Table 2. Summary of all tests: Dylos, Aerocet 531, Airbeam Just Fog, Airbeam JUUL and Airvisual JUUL.

Test Number	1		2		3		4		5	
µg/m^3	BAM 1020	Dylos	BAM 1020	Aerocet 531	BAM 1020	Airbeam Just Fog	BAM 1020	Airbeam JUUL	BAM 1020	Airvisual JUUL
Average bckg before e-cig	4.0	2.2	15.1	15.8	5.0	9.1	5.0	5.6	9.3	5.2
Average during e-cig	6.3	18.3	17.2	30.8	12.0	92.6	12.0	11.0	10.0	87.1
Average bckg after e-cig	4.8	1.6	16.7	22.8	11.0	10.5	13.0	8.0	9.6	5.3
Median during e-cig	5.5	23.0	17.5	35.5	12.0	97.3	12.0	29.5	10.0	691.2
Max during e-cig	8.0	38.7	19.0	55.1	12.0	188.4	12.0	55.0	10.0	1377.4
Min during e-cig	3.0	7.2	16.0	16.0	12.0	6.1	12.0	3.9	10.0	5.0

bckg = background.

Similar results were obtained when all other OPC models calibrated in SHS from CC were used in attempts to measure ECSHA.

As shown in Table S2, the Pearson correlations between all OPCs and the BAM and between each individual OPC are good, but the differences in the absolute concentration values differ so much that it becomes impossible to find the correct ECSHA density k factor for all OPCs when they must be employed in EC real-time environmental pollution aerosol measurements.

3.2. Measurements in Number of Particles

3.2.1. ECSHA Using "Elips C" ECs Measured in counts/cm^3: Model TSI 3007 with 2 min Sampling Time

The background measurements of the TSI 3007 started about 30 min before the EC test and ended only 10 min after because the instrument stopped working. As shown in Table S3 and Figure 3, counts/cm^3 increased from the initial background of 8109 to 14,296.

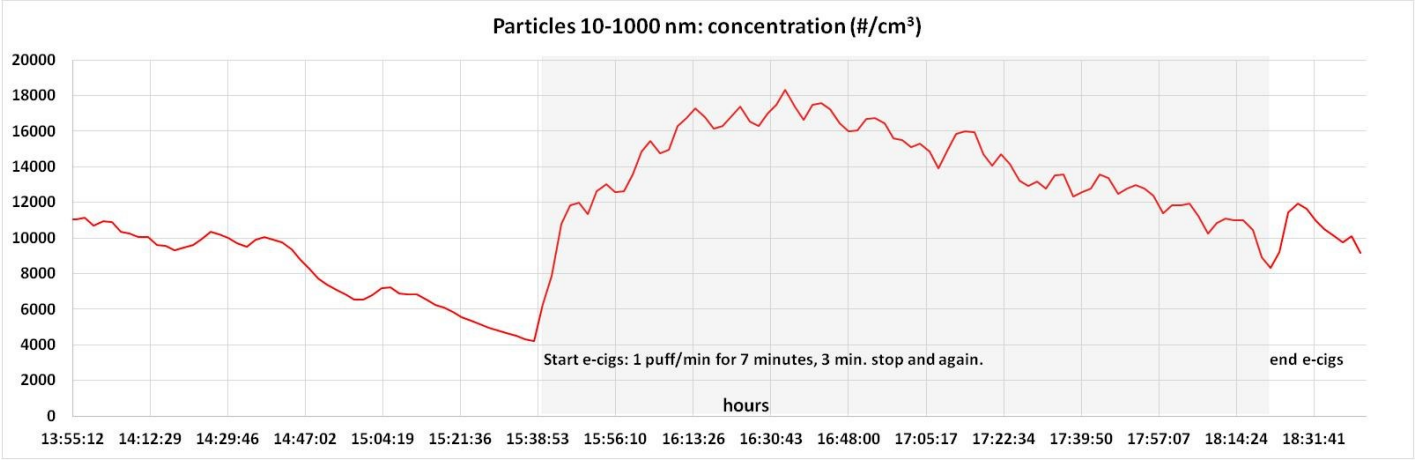


Figure 3. Real-time graph of ECSHA using "Elips C" ECs measured in counts/cm^3, with a sampling time of 2 min.

The background of 10,175 counts/cm^3 after EC use remained slightly higher than before, but this difference may be due to background changes not correlated with EC use and/or too short a measurement duration because of logistic problems. Large particles may have dropped, but small particles can linger for longer in this environment. Thus, this could be a real phenomenon associated with ultrafine particles below 100 nm in diameter.

3.2.2. ECSHA Using "Just Fog" ECs Measured in counts/L: Model Profiler 212-2 with 10 s Sampling Time

The 212-2 heater for automatic RH % control was switched off in all tests.

The background measurements started about one hour before EC use and ended one hour after.

With the 10 s time definition, it becomes possible to note the extreme volatility of the EC aerosol clouds after each e-cig puff. As shown in Table 3 and Figure 4, the Profiler's final background was slightly higher than the initial value, but this difference may be due to background changes not correlated with EC use and probably due to re-suspended particles from the presence of persons in the room.

Table 3. Details of ECSHA measured in counts/L with Profiler 212-2 when using "Just Fog" ECs.

Counts/L	0.3	0.5	0.7	1.0	2.5	3.0	5.0	10.0
Average bckg before ECs	27,045	2547	1034	657	192	101	27	6
Average during ECs	245,677	145,324	78,256	41,618	1947	306	28	7
Average bckg after e cigs	39,063	3132	1211	757	217	105	29	6
Median during EC	484,034	309,591	198,533	133,513	8082	1317	36	12
Max during ECs	911,826	597,365	387,585	262,971	15,892	2568	66	24
Min during ECs	56,241	21,817	9480	4054	271	66	6	0

Although the exponential equations representing the average background profile did not significantly differ from that of the average during the use of e-cig (Pearson R = 0.865), ECSHA showed a significant percentage reduction in the 0.4 µm size as compared with the background: 40.8 vs. 90.6%, but significant increases in other sizes (see Figure 5 and Table S4). The median particle size between >0.3 and >0.5 was 0.4 µmeters, and that between >5.0 and >10.0 was 7.5 µm.

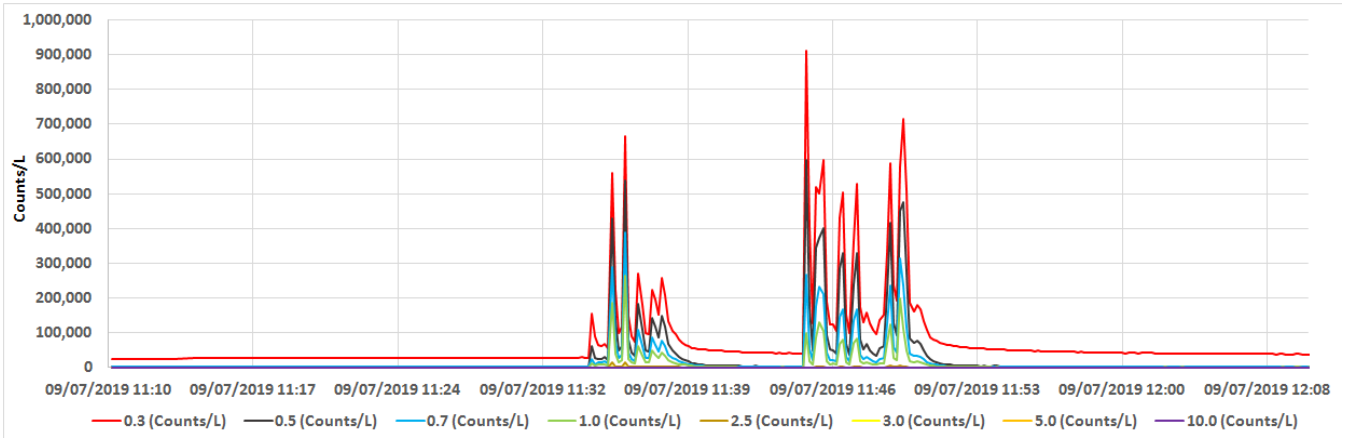


Figure 4. Real-time graph of ECSHA using "JustFog" ECs measured in counts/L, with a sampling time of 10 s.

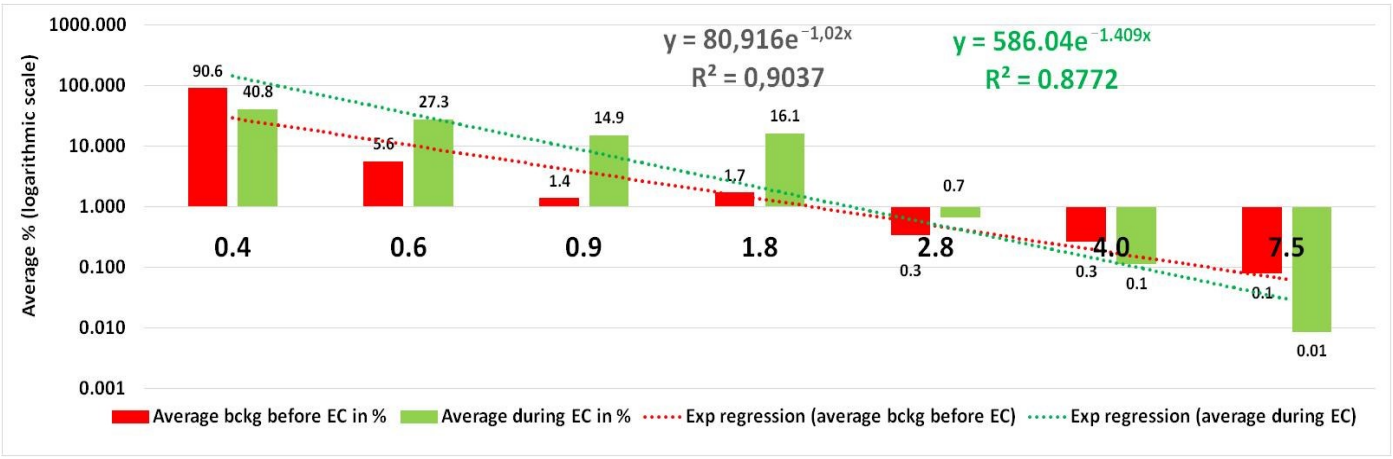


Figure 5. ECSHA profile comparison between background and during use of "JustFog" e-cigs.

3.2.3. ECSHA Using "JUUL" ECs Measured in counts/L: Model Profiler 212-2 with 10 s Sampling Time

The background measurements started about one hour before EC use and ended one hour after. As shown in Table 4 and Figure 6, also in this test, the Profiler's final background was slightly higher than the initial value, but this difference may be due to background changes not correlated with EC use.

Table 4. Details of ECSHA measured in counts/L with Profiler 212-2 when using "JUUL" ECs.

Counts/L	0.3	0.5	0.7	1.0	2.5	3.0	5.0	10.0
Average bckg before ECs	25,955	2005	714	436	119	62	14	2
Average during Ecs	107,984	38,608	14,542	6649	361	118	26	4
Average bckg after e cigs	26,489	1916	704	454	138	74	17	2
Median during EC	126,259	45,444	16,304	6954	355	120	27	6
Max during Ecs	204,102	79,904	27,958	11,543	499	168	48	12
Min during Ecs	48,415	10,983	4650	2364	211	72	6	0

Pearson between bckg before EC use and during EC use = 0.953. Wilcoxon Signed-Rank test (bckg before and during EC use) p value = 0.012.

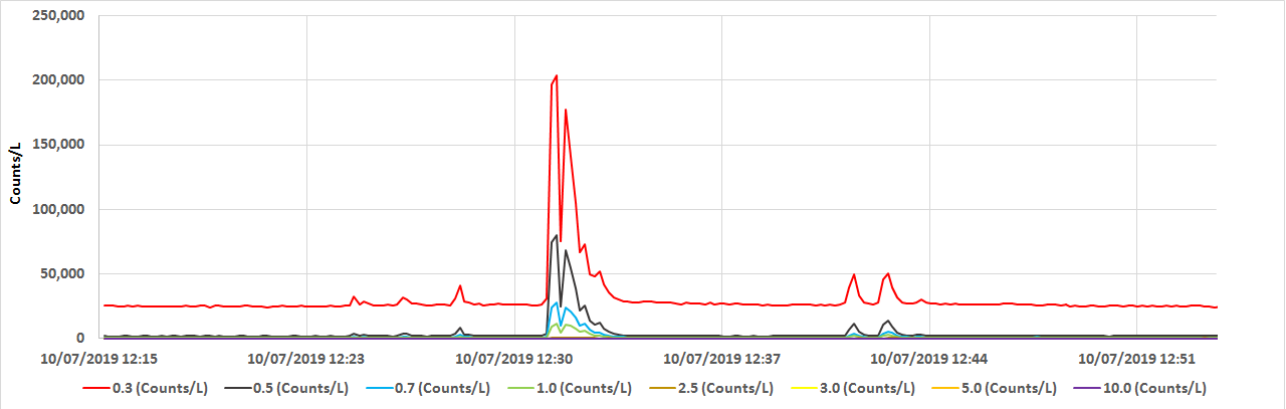


Figure 6. Real-time graph of ECSHA measured in counts/L using "JUUL" ECs, with a sampling time of 10 s.

Comparing ECSHA and background size profiles, we found a significant percentage increase in >0.5 (35.8 and 7.72%, respectively), >0.7 (13.5 and 2.75, respectively) and >1.0 (6.2 and 168, respectively). The Pearson correlation was good (R = 0.953), and the Wilcoxon test showed that the two distributions were significantly different in averages (p = 0.012). As shown in Figure 7 and Table S5, although the exponential equations representing the average background profile did not differ significantly from that of the average during the use of e-cig (Pearson R = 0.944), the ECSHA showed a significant percentage reduction in the 0.4 µm size as compared with the background: 62.1 vs. 92.3%, but significant increases of 24.1 vs. 5.0; 7.7 vs.1.1; and 5.7 vs. 1.2 in 0.6, 0.85 and 1.75 µm, respectively (see Table S6).

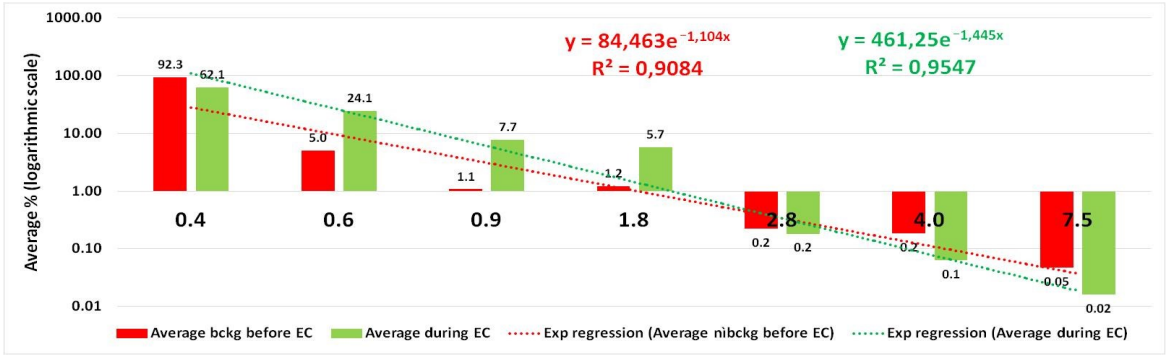


Figure 7. ECSHA profile comparison between background and during use of "JUUL" e-cigs.

The exponential ECSHA equations of the two brands were not significantly different, but JUUL showed a greater percentage of 0.4 and 0.6 µm and lower 0.85 and 2.75 µm percentages (see Figure 8).

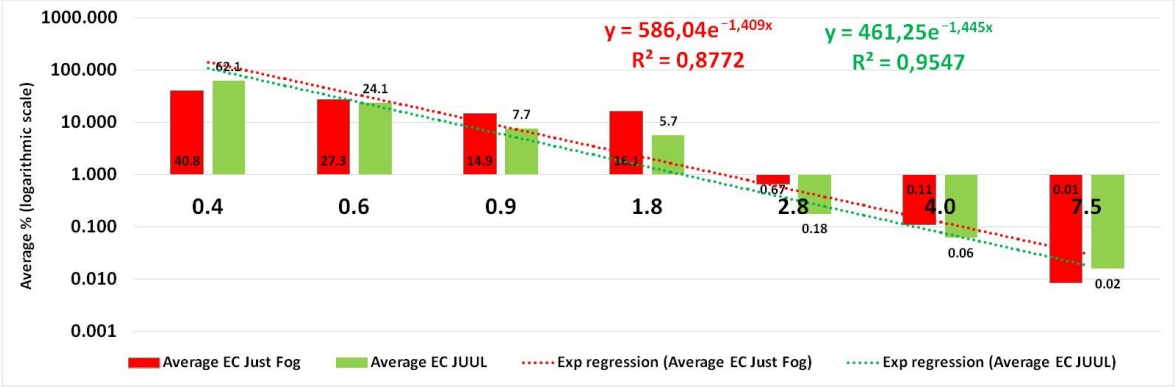


Figure 8. ECSHA profile comparison of JustFog and JUUL.

4. Discussion

4.1. Comments on Mass Measurements

In all ECSHA measurement tests, performed using four different OPC models pre-calibrated in mass in SHS from CC with different sampling times and three different brands of ECs, the results showed significant increases in $PM_{2.5}$ concentrations, while the reference instrument BAM-1020 indicated only small increments in all tests, except on the test of the CC where the BAM-1020 detected the PM (regression analysis: $y = 1.0703x - 16.743$, $R^2 = 0.9827$, see Figure 1). The significant increase in the OPC measurements is due to the fact that all OPCs detect volatiles, semi-volatiles, metals (K, Cr, Ni, Zn, Ag, La and Pb) and trace elements [50,51], while the BAM-1020 does not detect volatile and semi-volatile elements but detects metals and trace elements only because of RH control.

Additional difficulties arise because ECSHA is very unstable and subject to rapid changes from a liquid or semiliquid to the gaseous state [52] and because of the different modes of use of e-cigs by vapers (the possibility of selecting the vaporizer voltage, etc.). This fast change of state is largely correlated with environmental conditions such as T, RH, AER, etc., but can generally be evaluated in a few seconds, making it extremely difficult to reach the necessary uniform environmental aerosol distribution in all parts of the environment (mixing factor close to 1.0 on a scale from 0.0 to 1.0). As a consequence, it becomes almost impossible to simultaneously have the same concentration at the OPC to be calibrated and at the reference instrument sampling points.

Therefore, the two instruments will unavoidably measure different aerosol concentration levels, resulting in the impossibility of correctly calculating the density k factor.

The mixing factor on a scale from 0 to 1.0 is normally a factor indicating the spatial distribution of PM in a closed environment, such as a room or office. A value of 1.0 indicates that all PM sizes in all positions of the room are exactly at the same concentrations, and this is a necessary condition for the correct calibration of OPCs. Of course, this is not possible in a normal situation because of PM gravitational sedimentation (coarse PM sedimentation velocity is much faster than that of fine and ultrafine PM), and therefore, it is necessary to operate one or more fans to create strong turbulence in the air to distribute all PM sizes to all parts of the room volume.

4.2. Comments on Particle Number Measurements

Measurements performed using multiple-channel particle counters, such as the model 212-2 Profiler used in this study, will allow for a better understanding, in future research, of the lung deposition of inhaled ECSHA for each size of the particles and the evaluation of their deposition on the bronchial tree and small airways.

The measurement of the number of particles instead of mass could have some advantages; the most important are:

(a) No need to spend time on relatively complex calibration procedures to find the correct density k factor; normally, the accuracy is +/− 10% to calibrate aerosol (polystyrene latex spheres (PSLs));
(b) The possibility of establishing PM size profiles if multiple-channel OPCs are used. With these instruments, it is possible to compare PM size profiles before (the background), during and after the EC use phase.

Most OPCs do not measure particles smaller than 0.3 μm, and, as a consequence, measurements in mass of PM_1, $PM_{2.5}$ and PM_{10} miss these particles, which may not be significant in terms of mass but extremely important in terms of the number of particles because particle toxicity increases with decreasing particle diameter and increasing total particle surface area [53,54]. In addition, the deposition of particles within the alveolar regions of human lungs increases as the particle size decreases. Using CPCs such as the model TSI 3007 or equivalent to measure the number of particles of the 10 or even 5 nm size is highly recommended.

RH also interferes when the measurements are in number of particles because the diameter shows a tendency to shift to higher values as RH increases. The OPCs measure

the number of particles at different sizes. For each size, the volume is calculated, and by applying the density k factor, it is possible to calculate the mass. However, RH interference changes not only the density of the PM but also its volume, because particles soaked in water increase in volume [55] (see also Figures S1 and S2 in SI). Therefore, if the volume changes because of a change in RH, the size distribution also changes, introducing errors in the PM size profiles. However, if the environmental RH never exceeds 45/50%, as in this study, this shift probably does not produce significant differences in the size profiles of the PM (see Figure S3).

The extreme volatility of ECSHA causes the same detection problem as the measurements in mass, as described above.

Although, generally, the sample flow is within the range of a few liters/min, it is recommended that in all cases, the exhaust be directed, when possible, in the opposite direction of the sample inlet to avoid out/in recirculation, as well as in the case of the filtered clean air leaving the pump outlet. The results of these tests demonstrate that the measurement of environmental PM pollution generated by ECs using OPCs in real time is a real challenge for the following reasons:

(a) The extreme tendency of the aerosol clouds emitted by users of e-cig to evaporate;
(b) The impossibility of finding the proper density k factor to apply to the OPCs if the measurements are in mass.

However, a solution may be possible if the OPCs are individually pre-calibrated in mass for CCSHS or other reproducible PM compositions and the results are presented in ECSHA mass concentration in reference to CCSHS or to other selected and reproducible PM compositions. This method has been successfully applied in the measurements of Total Volatile Organic Compounds (TVOC) using the Flame Ionization and/or Photoionization Detection principle of operation, and the results are expressed in parts per million (ppm) in reference to isobutylene (mainly TVOC instruments are calibrated using this VOC) [56].

If multiple size channels are used, it becomes possible to elaborate the PM profile from 0.01 to 10.0 µm for a better understanding of the destiny of ECSHA when inhaled passively. Another suggestion is to reduce the sampling time, if possible, to one second for a better time definition. Although not solving the inaccuracy problems due to the extreme volatility of ECSHA, measurements in number of particles may be easier because they do not require calibration in mass since they are based on the manufacturer factory calibration, so the results are more reliable and repeatable by other studies.

5. Conclusions

In this study, we analyzed the problems encountered in providing measurements expressed as mass of EC emissions using OPC technology.

We found that a standard measurement requires the simultaneous operation of the OPC equipment and a certified reference instrument to detect density (k factor) for each specific PM source and different PM sizes.

Crucially, this setup may be inapplicable because the extreme volatility of EC-generated aerosol makes it impossible to gauge a correct EC k factor. Considering these limitations in the measurement of EC mass, we suggest that the measurement of EC number of particles may be more appropriate when using OPC technology.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/atmos13091393/s1. Figure S1: OPCs RH interference (Scattering Coefficient Ratio); Figure S2: OPCs RH interference Density k Factor Ratio; Figure S3: Example of Temperature (AT) and Relative Humidity (RH) during one test; Table S1: Details after calibration of OPC in SHS from CC; Table S2: Pearson correlation between all OPCs and the BAM and between each individual OPC; Table S3: Details of SHA measured in counts/cm^3 using "Elips C" ECs; Table S4: SHA profiles comparison between background and during use of "Just Fog" e-cig in percent; Table S5: SHA profiles comparison between background and during use of "JUUL" e-cig in percent; Table S6: SHA profiles comparison of Just Fog and JUUL in percent.

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