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Chlorination Cessation Alters Greenhouse Gas Dynamics in Artificial Urban Ponds

J. J. Montes-Pérez[1,2], P. Irusta[1], L. Cañas[1], F. Mejía[1,3], N. Pinaud-Brageot[4], B. Obrador[1,5], D. Puigserver[2,6], A. Millán[2,6], and D. von Schiller[1,2]

[1]Departament de Biologia Evolutiva, Ecologia i Ciències Ambientals, Universitat de Barcelona (UB), Barcelona, Spain, [2]Institut de Recerca de l'Aigua (IdRA), Universitat de Barcelona (UB), Barcelona, Spain, [3]Departamento de Ciencias de la Tierra y de la Construcción, Carrera de Ingeniería Civil, Universidad de las Fuerzas Armadas—ESPE, Sangolquí, Ecuador, [4]University of Poitiers, Poitiers, France, [5]Institut de Recerca de la Biodiversitat (IRBio), Universitat de Barcelona (UB), Barcelona, Spain, [6]Department of Mineralogy, Petrology and Applied Geology, Faculty of Earth Sciences, University of Barcelona (UB), Barcelona, Spain

Key Points:

- Chlorination cessation in artificial urban ponds increases CH_4, lowers N_2O, and has no effect on CO_2 emissions
- In CO_2 equivalents, non-chlorinated, and chlorinated ponds show similar greenhouse gas emissions
- Pond naturalization measures can enhance urban resilience without notably raising greenhouse gas emissions

Supporting Information:

Supporting Information may be found in the online version of this article.

Correspondence to:

J. J. Montes-Pérez,
jmontesp@ub.edu

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Author Contributions:

Conceptualization: J. J. Montes-Pérez, B. Obrador, D. von Schiller
Data curation: J. J. Montes-Pérez
Formal analysis: J. J. Montes-Pérez, D. von Schiller
Funding acquisition: D. von Schiller
Investigation: J. J. Montes-Pérez, P. Irusta, L. Cañas, F. Mejía, N. Pinaud-Brageot, B. Obrador, D. Puigserver, A. Millán, D. von Schiller
Methodology: J. J. Montes-Pérez, L. Cañas, B. Obrador, D. von Schiller
Project administration: D. von Schiller
Supervision: B. Obrador, D. von Schiller
Visualization: J. J. Montes-Pérez, D. von Schiller

Abstract Cities are facing an ecological challenge, and international policies are increasingly focused on implementing nature-based solutions to support this transition. In this context, the naturalization of artificial urban ponds (AUP) is a promising approach with proved benefits for biodiversity and human well-being. However, the naturalization of AUP may be accompanied by increased greenhouse gas (GHG) emissions. Here, we evaluated the effect of chlorination cessation, an essential step in the naturalization process, on GHG dynamics in AUP. Partial pressures of CO_2 (pCO_2), CH_4 (pCH_4), and N_2O (pN_2O) were measured in 41 artificial urban ponds (28 non-chlorinated and 13 chlorinated) in the city of Barcelona during winter and summer to assess: (a) the effect of chlorination treatment, (b) the effect of seasonality, and (c) the main drivers behind the partial pressures of these GHGs. Results show that although chlorination cessation increased pCH_4, it reduced pN_2O and had no significant effect on pCO_2. The main drivers of these patterns were naturalization, with factors related to primary production playing a major role; seasonality, with temperature as a key environmental variable; and groundwater legacy. Importantly, the net global warming potential (GWP), expressed as CO_2 equivalents, was not significantly higher in non-chlorinated ponds. These findings suggest that the naturalization of artificial water bodies could be a viable strategy to create more resilient cities without significantly increasing GHG emissions.

Plain Language Summary Cities have a big challenge trying to be more ecologically sustainable. In this context, naturalization of artificial urban ponds (AUP) is a way to increase biodiversity and human well-being. However, when chlorination is stopped in AUP, the conditions for the natural production of CO_2, CH_4, and N_2O, the three most important greenhouse gases may be created. This could be a negative and undesirable side effect. The objective of this work was to evaluate the magnitude of this effect. To do that, we measured the concentration of CO_2, CH_4, and N_2O in 41 AUP (28 non-chlorinated and 13 chlorinated) in the city of Barcelona in summer and in winter. We did not find significant differences in the concentration of CO_2 between non-chlorinated and chlorinated ponds. However, in non-chlorinated AUP, the concentration of CH_4 was higher, whereas the N_2O concentration was lower. Despite these differences, if we sum up the three greenhouse gases as CO_2 equivalents (which represents the net global warming potential) and estimate the potential emissions, there were no significant differences between the emissions from non-chlorinated and chlorinated AUP. Therefore, these results suggest that we could naturalize AUP, obtaining their proven benefits, without significantly increasing greenhouse gas emissions.

1. Introduction

Cities host more than half of the world's population and are responsible for up to 70% of global greenhouse gas (GHG) emissions (IPCC, 2021). Consequently, cities face a critical challenge in achieving environmental sustainability. In fact, international agreements, such as the European Green Deal, the Glasgow Pact, and the Paris Agreement urge countries to commit to reducing their GHG emissions and achieving net-zero emissions by 2050. In this context, the implementation of nature-based solutions (NbS) has been proposed as a key action to mitigate emissions and transition toward more sustainable cities (Kabisch et al., 2017).

Writing – original draft: J. J. Montes-Pérez
Writing – review & editing: B. Obrador, D. Puigserver, D. von Schiller

Among NbS, the naturalization of urban spaces is considered an effective tool for carbon sequestration and reduction of GHG emissions (Pan et al., 2023), helping to reach the sustainable development goals outlined in the United Nations' Agenda 2030 (United Nations, 2015). Although the implementation of NbS in terrestrial ecosystems, such as parks or urban forests, have demonstrated significant capacity for reducing GHG emissions (Nowak et al., 2013; Zhang et al., 2022; Zhao et a., 2023), the impact of naturalization of urban aquatic ecosystems on GHG fluxes remains poorly understood.

In many cities, artificial water bodies are constructed primarily for esthetic purposes, adorning parks or gardens. These water bodies are often treated with chlorine to prevent algal growth. However, the naturalization of these water bodies, through the redesign of physical, chemical, and biological components, represents a NbS that could be an effective strategy to improve the ecosystem services they provide, including human well-being (Oertli et al., 2023). A key first step in this process is the cessation of chlorine use, which can then be followed by additional measures such as the introduction of aquatic vegetation and the creation of irregular, shallow shorelines to increase habitat complexity and biodiversity. When the chlorination treatment ceases, artificial urban ponds (AUP) may develop ecosystem functions similar to those of natural ponds, which can act as carbon sinks (Downing et al., 2008; Taylor et al., 2019) but, at the same time, can also be a significant source of GHGs (Malyan et al., 2022; Ray & Holgerson, 2023).

Recently, increasing attention has been given to the role that naturalized urban aquatic ecosystems can play in regulating GHG fluxes. Several studies have revealed that naturalized urban ponds tend to be supersaturated with methane (CH_4) and carbon dioxide (CO_2) (Audet et al., 2020; Bauduin et al., 2024a, 2024b; Peacock et al., 2021; Ray & Holgerson, 2023; van Bergen et al., 2019). The concentration of CH_4 and CO_2 may vary according to seasonal (Jensen et al., 2022; Ray et al., 2023; van Bergen et al., 2019) and diel (Natchimuthu et al., 2014; Sand-Jensen et al., 2019) changes in environmental variables. Nevertheless, further research is needed to increase our understanding on how naturalization can alter the processes controlling CO_2 and CH_4 emissions in urban ponds. In addition, most studies on nitrous oxide (N_2O) dynamics in urban ponds show that they can be also a source of N_2O, but with a relatively smaller contribution compared to other GHGs (Audet et al., 2020; Bauduin et al., 2024a, 2024b; Ray & Holgerson, 2023). Supersaturated N_2O concentrations are attributed to processes such as nitrification and denitrification (Pan et al., 2023; Pauleta et al., 2019) and, therefore, can be potentially influenced by naturalization. Furthermore, measurements of GHG concentrations in ponds are typically based on daytime measurements, which may introduce biases by either underestimating or overestimating emissions (Sø et al., 2024). This diel effect could be strongly amplified by naturalization, although it remains uncertain.

Here, we aimed to evaluate the effect of chlorination cessation, an essential step in the naturalization process, on GHG dynamics in AUP. With this purpose, we quantified the partial pressures of CO_2, CH_4, and N_2O across a wide range of AUP in the city of Barcelona, Spain. To explore seasonal and diel variability in GHG fluxes, along with potential drivers, we conducted samplings during two contrasting seasons (summer and winter) and times of the day (daytime and nighttime). We hypothesized that non-chlorinated AUP would exhibit higher GHG partial pressures due to increased biological activity. We further hypothesized that in non-chlorinated AUP, metabolic processes would be more intense in summer, resulting in higher GHG partial pressures compared to winter, whereas no seasonal differences would exist in chlorinated AUP. Finally, we hypothesized that in non-chlorinated AUP, daily changes in photosynthetic activity would directly influence pCO_2 and indirectly other metabolic processes, that is, methanogenesis and nitrification/denitrification, which produce CH_4 and N_2O. Therefore, we predicted significant changes in GHG partial pressures across seasons and between day and night in non-chlorinated AUP, and no seasonal or daily variations in chlorinated AUP. As a final step, we explored the effect of chlorination cessation on the potential diffusive fluxes in terms of global warming potential (GWP) that could be emitted from the studied AUP.

2. Materials and Methods

2.1. Study Area and Sampling Design

Barcelona, a city in northeastern Spain with approximately 1.6 million inhabitants, experiences a typical Mediterranean climate characterized by hot, dry summers and mild, wet winters. For this study, we selected 41 AUP distributed across the city, categorized into two types: 13 chlorinated (i.e., subject to chlorine treatment) and 28 non-chlorinated (i.e., without chlorine treatment) (Figure 1). In this study, chlorination cessation is used as a proxy of naturalization of an AUP. Although not in all cases, removal of chlorine treatment is typically

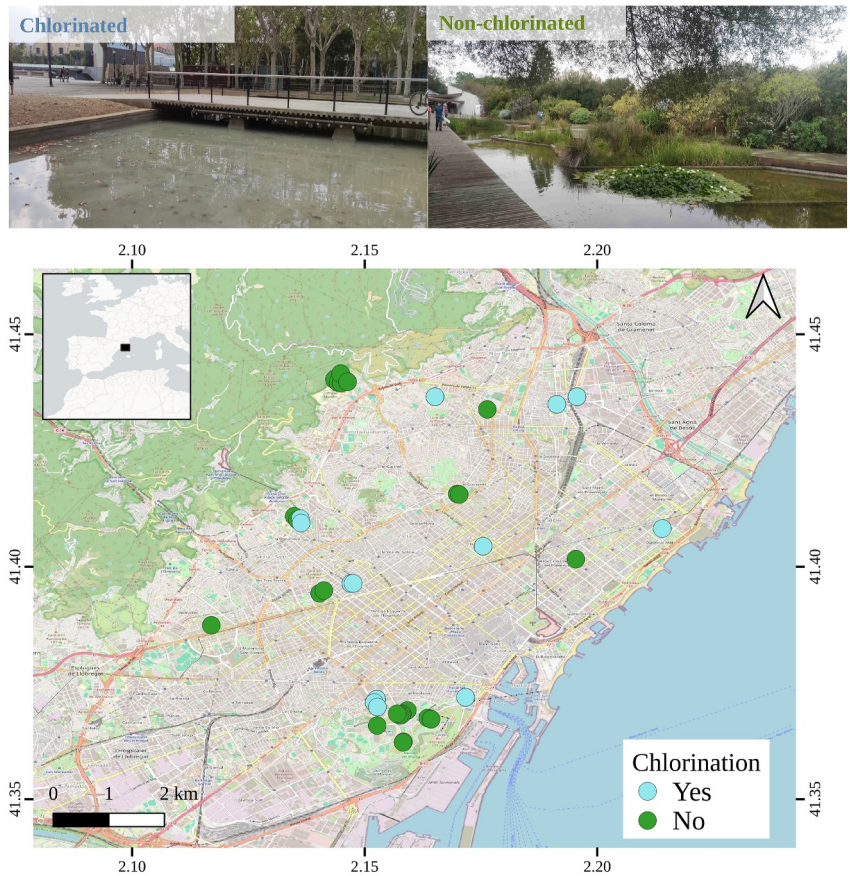

Figure 1. Top: Example images of chlorinated (left) and non-chlorinated (right) urban ponds. Bottom: Map of Barcelona showing the locations of the sampled chlorinated (n = 13) and non-chlorinated (n = 28) urban ponds. The inset shows the location of Barcelona within the Iberian Peninsula. Coordinates are expressed in decimal degrees. Base map and data from OpenStreetMap and OpenStreetMap Foundation (CC-BY-SA). © https://www.openstreetmap.org and contributors.

complemented by the introduction of aquatic vegetation, such as emergent, submerged, or floating species, though these measures are not applied consistently across all sites. This criterion was used to follow the guidelines set by the Barcelona City Council, where they remove chlorination to ensure the success of vegetation introduction, allow other organisms to colonize the AUP, and increase biodiversity. Information regarding the chlorine treatment of AUP was provided by the Barcelona City Council and corroborated by our own physicochemical analyses. All selected ponds were artificial, with concrete bottoms. However, many AUP, especially the non-chlorinated ones, contained varying proportions of introduced vegetation, including submerged, emergent, and floating vegetation, as well as the necessary planting beds to support this vegetation (Figure 1). The water sources for these ponds are diverse and uncertain, as they are refilled with groundwater from different wells, municipal water, or a mixture, depending on availability. The sampled AUP varied in surface area from 5 to 2,225 m^2 and in volume from 3 to 3,500 m^3. Additional information about physicochemical variables and characteristics of all sampled AUP can be found in Table S1 in Supporting Information S1.

To explore seasonal variability, sampling was conducted in two contrasted seasons: summer (24–28 July 2023) and winter (2–25 January 2024). Here, we selected summer and winter as representative of the two extremes of annual variability. Summer is characterized by the highest temperatures, potential stratification, and high primary production activity, whereas winter features the lowest temperatures, a well-mixed water column, and low primary production. In each season, all AUP were sampled on a specific daytime window: from 17:00 to 20:30 in summer, and from 14:00 to 17:30 in winter.

To assess diel variability, we selected eight chlorinated and eight non-chlorinated AUP for additional nighttime sampling. Nighttime sampling was carried out before sunrise, from 03:00 to 06:00 in summer, and from 04:00 to 07:00 in winter, to capture maximum diel variation (Ran et al., 2022; Taylor et al., 2024).

We evaluated the short-term temporal variability in GHG partial pressures to rule out potential biases from a 1-week sampling period. During the winter sampling, we selected 7 AUP for repeated sampling 1 week later. We used paired Student's *t*-tests to analyze differences in the partial pressure of CO_2 (pCO_2), CH_4 (pCH_4), and N_2O (pN_2O) between the two sampling periods. We found no significant differences for pCO_2 ($t(6) = 0.29$, $p = 0.78$), pCH_4 ($t(6) = -1.32$, $p = 0.24$), or pN_2O ($t(6) = -1.35$, $p = 0.23$).

To assess the potential contribution of GHG from groundwater, GHG partial pressures were measured at 7 groundwater extraction points supplying water to studied AUP in Barcelona during September 2023 and January 2024.

2.2. Field Sampling

Temperature, pH, conductivity, and dissolved oxygen were measured in situ below the water surface (~5 cm) using a multisonde WTW pH/Cond 3320 2EA310 and an oximeter YSI ProODO. Percentage cover of floating (e.g., Nymphaea sp.), emerged (e.g., Juncus sp.), and submerged (e.g., Chara sp.) vegetation was visually estimated in situ.

To measure pCO_2, pCH_4, and pN_2O in the water, three subsamples were taken at each pond below water surface using the headspace technique (Striegl et al., 2012). A syringe equipped with a three-way valve was used to extract 30 mL of water, followed by an additional 30 mL of air to create the headspace. To ensure rapid equilibrium between the water and air interface, the syringes were vigorously shaken for 1 min and then maintained at a constant temperature for 2–3 min. Subsequently, 20 mL of the equilibrated gas was injected into a 12 mL Exetainer® vial. The volume injected (20 mL) into the vial exceeded that of the vial to create an overpressure within the vial, thus safeguarding against contamination or dilution by external air. The samples were stored at 4°C until analysis. For the determination of dissolved organic carbon (DOC), total dissolved nitrogen (TDN), total phosphorus (TP), anions, major, and minor elements, and pigments, three subsamples of 500 mL of water were collected from each pond below water surface, stored in a portable fridge and transported to the laboratory for further processing and analysis.

2.3. Laboratory Analyses

To determine the concentration of DOC and TDN, 45 mL of each sample were filtered using pre-rinsed sterile Whatman FP cellulose acetate microfiber filters with a pore diameter of 0.45 µm. Filtered samples were acidified to a pH of 2 using a 10% HCl solution. The samples were stored at 4°C until analysis. DOC and TDN concentration were measured with a Shimadzu TOC-VCSH equipped with a TNM-1 module.

Water samples for TP were immediately frozen and stored at −20°C until analysis. Subsequently, 20 mL of unfiltered sample was mixed with 2 mL of oxidative reagent and autoclaved at 110°C for 90 min (Grasshoff, 1983). Soluble reactive phosphorus was then measured using the ascorbic acid method (Murphy & Riley, 1962), with absorbance read at 880 nm using a Shimadzu Pharmaspec UV-1700 spectrophotometer.

Major elements (K, Mg, Ca, Na, P, S, and Si) and minor elements (Sr, Al, Ba, and Cu) were analyzed using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) with a Perkin Elmer Optima 8300 spectrometer. Additionally, due to their low concentrations, Mn and Fe were analyzed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) with a Perkin Elmer NexIon 2000 spectrometer. For these analyses, water samples were filtered through Whatman FP cellulose acetate microfiber filters with a pore size of 0.45 µm, acidified with 65% HNO_3, and stored at 4°C until analysis. Anions (Cl^-, SO_4^{2-}, NO_3^-, NO_2^-, PO_4^{3-}) were analyzed using ion chromatography with a Jasco liquid chromatograph: Jasco PU-2089 plus pump, Jasco AS-2055 plus injector, Jasco W-2075 plus UV-Vis detector. Water samples were filtered through Whatman FP cellulose acetate microfiber filters with a pore size of 0.45 µm and stored at −20°C until analysis.

To determine the concentrations of chlorophyll-*a* and pheopigments, 300 mL of water were filtered through a GF/F glass microfiber filter. Subsequently, filters were wrapped in aluminum foil and stored at −20°C until further analysis. Pigments were extracted using 90% alkaline acetone and quantified using a Shimadzu Pharmaspec

UV-1700 spectrophotometer. Following the initial measurement, samples were acidified with 1N HCl and absorbance remeasured (Wetzel & Likens, 2000). The concentrations were calculated using the equations provided by Lorenzen (1967).

The concentrations of CO_2, CH_4, and N_2O in the headspace equilibrated gas were measured using an Agilent 7820A gas chromatograph equipped with Flame Ionization (FID) and Electron Capture (ECD) detectors. Calibration curves were done from a certificated standard gas mixture (Air liquid) with concentration 3,000, 15, and 6 ppm for CO_2, CH_4, and N_2O, respectively. The concentrations outside this range had to be extrapolated (ca. 7% for CO_2, 30% CH_4, and 15% for N_2O). Aqueous CO_2, CH_4, and N_2O concentration at field conditions were determined from measured headspace gas volume fractions and concentrations, based on the barometric pressure at the sampling site, field water temperature, equilibration temperature, and the corresponding Henry's law constants (Sander, 2015).

Subsequently, aqueous gas concentrations ($\mu mol \cdot L^{-1}$) were converted into partial pressures (µatm) using Henry's law:

$$pGHG = \frac{[GHG]}{k_{h,GHG}}$$

where pGHG is the gas partial pressure (CO_2, CH_4 or N_2O), in µatm, [GHG] is the concentration of the gas in the water, in $\mu mol \cdot L^{-1}$, and $k_{h,GHG}$ is the solubility constant for the specific gas at the sample water temperature, expressed in $mol \cdot L^{-1}\ atm^{-1}$.

2.4. Estimation of Diffusive GHG Fluxes

We estimated the potential diffusive GHG flux for each gas based on the respective gas exchange coefficient and the concentration gradient between the actual pond water and its theoretical equilibrium concentration with the atmosphere. This estimation was performed using the following equation:

$$F = k(GHG_{water} - GHG_{atm})$$

where F represents the estimated diffusive flux ($mmol \cdot m^{-2}\ d^{-1}$), k is the gas exchange coefficient for the specific GHG ($m \cdot d^{-1}$), and GHG_{water} and GHG_{atm} are the concentrations ($mmol \cdot m^{-3}$) of the gas in water and the concentration in equilibrium with the atmosphere, respectively.

The gas exchange coefficient was calculated based on the Schmidt number for each gas, using the equation proposed by Wanninkhof (2014), and the gas transfer velocity (k_{600}). Although there is a predictable relationship between wind speed and k_{600} at low-wind conditions, this relationship is largely independent (Cole & Caraco, 1998; Clark et al., 1995; Read et al., 2011). Such low-wind conditions are common in small, sheltered water bodies such as urban ponds, where convection tends to dominate turbulence; but there is still considerable uncertainty associated with estimating fluxes from convection (Read et al., 2011).

This uncertainty makes k_{600} estimations highly variable. Therefore, we adopted a conservative approach by applying a fixed gas transfer velocity for each pond size class: for AUP < 1,000 m^2, we used $0.3\ m \cdot d^{-1}$ and for AUP > 1,000 m^2, we applied $0.48\ m \cdot d^{-1}$, as implemented in similar studies (Holgerson & Raymond, 2016; Peacock et al., 2019). Certainly, this approach may introduce some uncertainty in the absolute magnitude of the estimated diffusive fluxes. However, the use of a chosen k_{600} affects the estimates of all three GHGs equally, preserving the relative contribution of each gas. Moreover, the studied AUP are small and located within a localized geographical area, experiencing similar meteorological conditions (e.g., temperature, solar radiation, and wind). Sampling was also conducted over a short time frame. As a result, variability in k_{600} is expected to be comparable between chlorinated and non-chlorinated AUP, minimizing potential bias in diffusive flux estimates between these groups due to differences in k_{600}.

To compare the GWP between studied gases, the diffusive flux of CH_4 and N_2O, were expressed as CO_2 equivalents (CO_2eq) following the emission metric proposed in AR6 of IPCC for a 100-year horizon (IPCC, 2023). The values of 27 for CH_4 and 273 for N_2O were applied.

2.5. Statistical Analysis

Principal components analysis (PCA) was used to reduce dimensionality and explore potential clustering of the ponds based on their environmental variables. The variables included in the PCA are detailed in Table S2 in Supporting Information S1.

A two-way mixed analysis of variance (ANOVA) with one between-subjects factor (Chlorination: Yes vs. No) and one within-subjects factor (Season: winter vs. summer) was used to test the effects of chlorination cessation and seasonality. To examine the differences between day and night, a paired *t*-test was employed for each Chlorination and Season factor combination, with the Wilcoxon rank-sum test used as an alternative when the assumptions for the *t*-test were not met. A Holm-Bonferroni correction was applied to minimize type I error (Aickin & Gensler, 1996). To avoid interference from the day-night cycle, only values obtained during daytime sampling were included for the two-way ANOVA. When the interaction was significant, Tukey's test was used to determine pairwise differences between means.

Prior to analysis, data were log-transformed to meet statistical assumptions (normality, homogeneity of variance, and homogeneity of covariances). These assumptions were assessed using the Shapiro-Wilk test, Levene's test, and Box's *M* test, respectively. All statistical analyses were conducted using the *R* package "rstatix" (Kassambara, 2023). When samples did not meet homoscedasticity criteria, as an alternative to the two-way mixed ANOVA, a linear mixed model (LMM) was applied to evaluate the significance of fixed effects (Chlorination and Season), with AUP treated as random effect. *F*-tests of fixed terms and degrees of freedom were calculated using the Kenward-Roger approximation (Luke, 2017) via the "lmerTest" *R* package (Kuznetsova et al., 2017). In such cases, differences between groups were calculated using the Estimated Marginal Means (EMMs) function provided by the "emmeans" package (Lenth et al., 2024). When reporting ANOVA and LMM results, the following format is used: F (between groups df, within groups df) = [F-value], p = [p-value]. For Tukey and EMM post hoc tests, results are reported as p = [p-value], 95% C.I. = [lower, upper].

To identify potential variables controlling the partial pressure of each studied GHG, multiple linear regression was applied, with variable selection performed using stepwise selection based on the Akaike Information Criterion (AIC). Nonsignificant variables were removed from the model following Crawley (2012). Model selection and subsequent trimming were conducted using the "stats" and "fuzzySim" *R* packages (Barbosa et al., 2024; R Core Team, 2024). To minimize multicollinearity, 17 variables were selected (Table S2 in Supporting Information S1) based on a correlation threshold of less than 0.7 between variables (Dormann et al., 2013) and a variance inflation factor (VIF) below 3 (Zuur et al., 2010). For all three models, linearity, error distribution, normality of residuals, homogeneity of variance, and extreme values (e.g., Cook's distance) were visually inspected (Figures S1–S3 in Supporting Information S1).

3. Results

3.1. AUP Features

The PCA showed considerable overlap between chlorinated and non-chlorinated AUP (Figure 2). Nevertheless, partial spatial segregation between the pond types was evident along the first principal component. This separation was mainly driven by differences in chemical variables, including conductivity, Cl, Na, Ca, NO_3^-, TDN, TP, DOC, and pH, all of which differed significantly between the groups (see Table S1 in Supporting Information S1). Significant differences were also noted in biological variables, such as chlorophyll-*a* concentration and percentage pond surface covered by vegetation. Overall, non-chlorinated AUP showed lower nutrient concentrations as well as higher pH and a higher presence of vegetation compared to chlorinated AUP.

3.2. GHG Partial Pressures

Pond water was generally supersaturated with respect to atmosphere for the three measured GHGs (Figure 3 and Table S3 in Supporting Information S1). For both pond types (chlorinated and non-chlorinated) and seasons (summer and winter), the median pCO_2 was 1,232 ($Q1$ = 753, $Q3$ = 2,784) µatm, pCH_4 was 188 (23, 719) µatm, and pN_2O was 1.66 (0.57, 3.46) µatm. However, the effect of chlorination and season differed among GHGs.

The pCO_2 ranged from 150 µatm (chlorinated pond "Parc Diagonal" in winter), to 22,000 µatm (non-chlorinated pond "Cascades1" in summer). However, no significant differences were found between chlorinated and

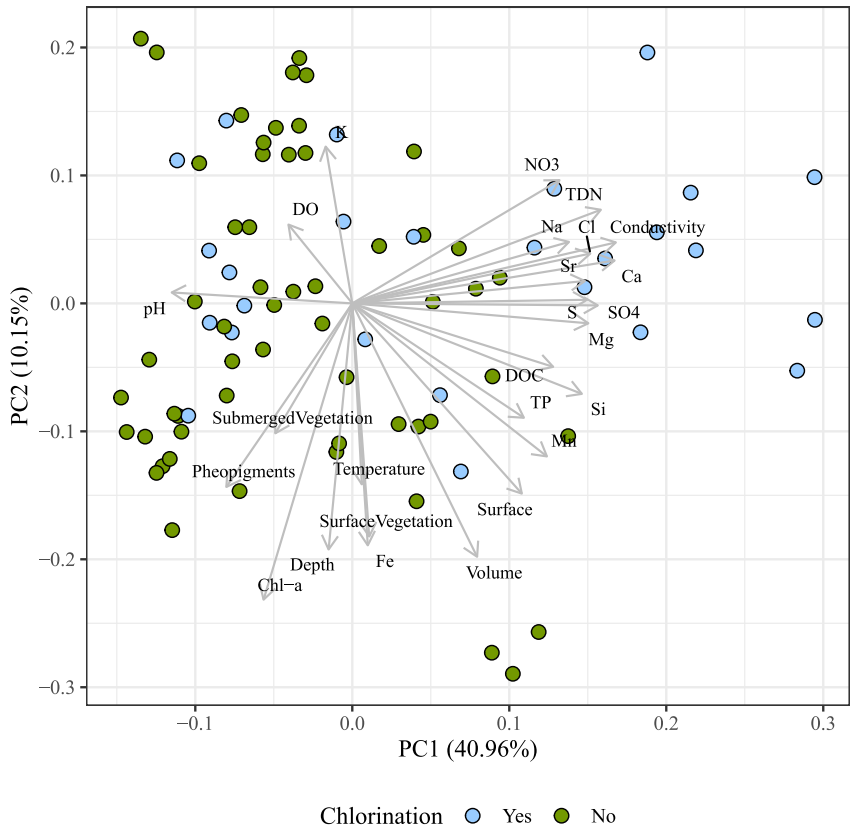

Figure 2. Principal component analysis of chlorinated (blue) and non-chlorinated (green) urban ponds based on physico-chemical and biological variables. Data from both winter and summer seasons are included. Full definition of the abbreviations of variables can be found in Table 2 in Supporting Information S1.

non-chlorinated AUP ($F(1, 38) = 0.57$, $p = 0.45$), nor was there a significant difference between pCO_2 measured in winter and summer ($F(1, 38) = 4.05$, $p = 0.05$). No interaction effect was found either ($F(1, 38) = 0.11$, $p = 0.74$).

The pCH_4 ranged from 8 µatm in the non-chlorinated pond "Estany Superior" in winter to 16,245 µatm in the non-chlorinated pond "Font Nimfa" in summer. Non-chlorinated AUP showed higher pCH_4 with respect to chlorinated AUP in both winter and summer ($F(1, 39) = 17.99$, $p < 0.001$). In addition, a significant effect of season was found ($F(1, 39) = 46.65$, $p < 0.001$), as well as an interaction between season and chlorination ($F(1, 39) = 6.63$, $p = 0.01$). In non-chlorinated AUP, pCH_4 was significantly higher in summer than in winter ($p < 0.001$, 95% CI = [−3.16, −1.04]), whereas this seasonal difference was not observed in chlorinated AUP ($p = 0.38$, 95% CI = [−2.5, 0.6]).

The pN_2O ranged from 0.06 µatm in the non-chlorinated pond "Aclimatació" in summer to 242 µatm in the chlorinated pond "Font Triangular" in summer. Both chlorination ($F(1, 39) = 21.53$, $p < 0.001$) and season ($F(1, 39) = 10.37$, $p = 0.003$) had a significant effect on pN_2O. Moreover, the interaction between these factors was also significant ($F(1, 39) = 12.44$, $p = 0.001$). Non-chlorinated AUP showed lower pN_2O than chlorinated AUP during summer ($p < 0.001$, 95% CI = [1.39, 3.78]), but no significant differences between pond types were observed in winter ($p = 0.07$, 95% CI = [−0.07, 2.31]). In the same line, the effect of seasonality was only significant in non-chlorinated AUP ($p < 0.001$, 95% CI = [−2.02, −0.77]), with lower pN_2O in summer.

3.3. Diel Changes

There were no significant differences between day and night in the partial pressure of any of the three studied gases (CO_2, CH_4, and N_2O) (Figure 4). This lack of diel changes was consistent across chlorinated and non-chlorinated AUP, as well as during summer and winter.

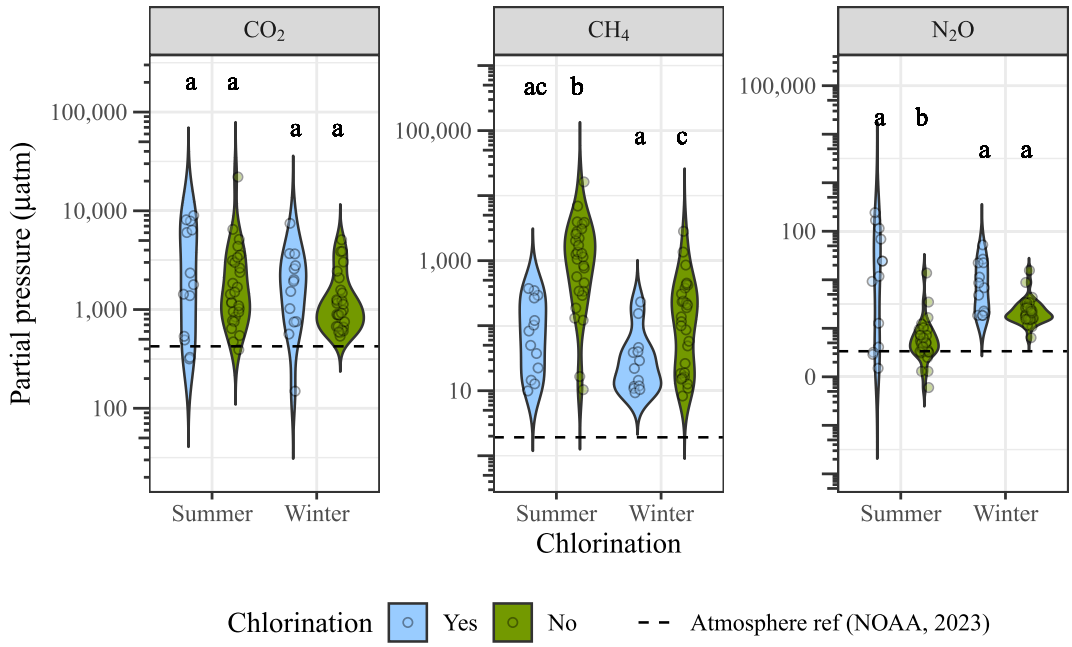

Figure 3. Density plot of partial pressures (µatm) for CO_2, CH_4 and N_2O in chlorinated (blue) and non-chlorinated (green) urban ponds measured during summer and winter. Points represent individual measurements. The dashed line indicates the reference value of partial pressure in the atmosphere according to NOAA 2023 (https://gml.noaa.gov/ccgg/trends/). Letters indicate statistical comparisons between groups within each gas. Groups sharing the same letter within a gas are not significantly different from each other, whereas groups with different letters within a facet have significant differences. Note log-scale in the *y*-axis.

3.4. Drivers

The multiple linear regression models obtained explained a large part of the variability in pCO_2 ($R^2adj = 0.62$), pCH_4 ($R^2adj = 0.70$), and pN_2O ($R^2adj = 0.72$) (Table 1).

Water temperature showed a positive relationship with pCO_2, suggesting that higher temperatures lead to increased pCO_2. Additionally, pCO_2 exhibited a negative relationship with dissolved oxygen, indicating that lower oxygen availability is associated with higher CO_2 levels. Mn was positively correlated with pCO_2, whereas Fe and pH showed negative correlations. Surface vegetation cover also positively influenced pCO_2, with more vegetated AUP showing higher CO_2 levels. Water temperature also showed a positive relationship with pCH_4. In addition, a negative relationship with SO_4^{2-} and dissolved oxygen suggests that AUP with lower concentration of these variables had higher pCH_4. Chlorophyll-*a* and submerged vegetation cover positively correlated with pCH_4.

Water temperature also showed a relationship with pN_2O, but in this case, it was negative. Thus, lower temperatures favored higher pN_2O, unlike the trends seen for CO_2 and CH_4. Similar to pCO_2, pN_2O was negatively associated with SO_4^{2-}, Fe, and pH and positively correlated with Mn and NO_3^-. Additionally, pN_2O showed a positive relationship with submerged vegetation cover.

3.5. Diffusive Fluxes

The estimated diffusive fluxes indicated that most studied AUP acted as sources of GHGs, regardless of whether they were chlorinated or non-chlorinated (Figure 5 and Table S4 in Supporting Information S1). The total flux ($CO_2 + CH_4 + N_2O$) in terms of CO_2 equivalents did not show significant differences between chlorinated AUP, 4,022 (479, 7,033) mg $CO_2eq \cdot m^{-2} \cdot d^{-1}$, and non-chlorinated AUP, 1,012 (694, 1,738) mg $CO_2eq \cdot m^{-2} \cdot d^{-1}$ ($W = 229$, $p = 0.2$). However, significant differences in the magnitude and relative contribution of fluxes were found between chlorinated and non-chlorinated AUP when comparing each of the gases studied.

The CO_2 flux in non-chlorinated AUP [537 (271, 1,042) mg $CO_2eq \cdot m^{-2} \cdot d^{-1}$], represented ~70% of the total flux from these ponds. In chlorinated AUP, the CO_2 flux [1,047 (340, 2,888) mg $CO_2eq \cdot m^{-2} \cdot d^{-1}$] accounted for

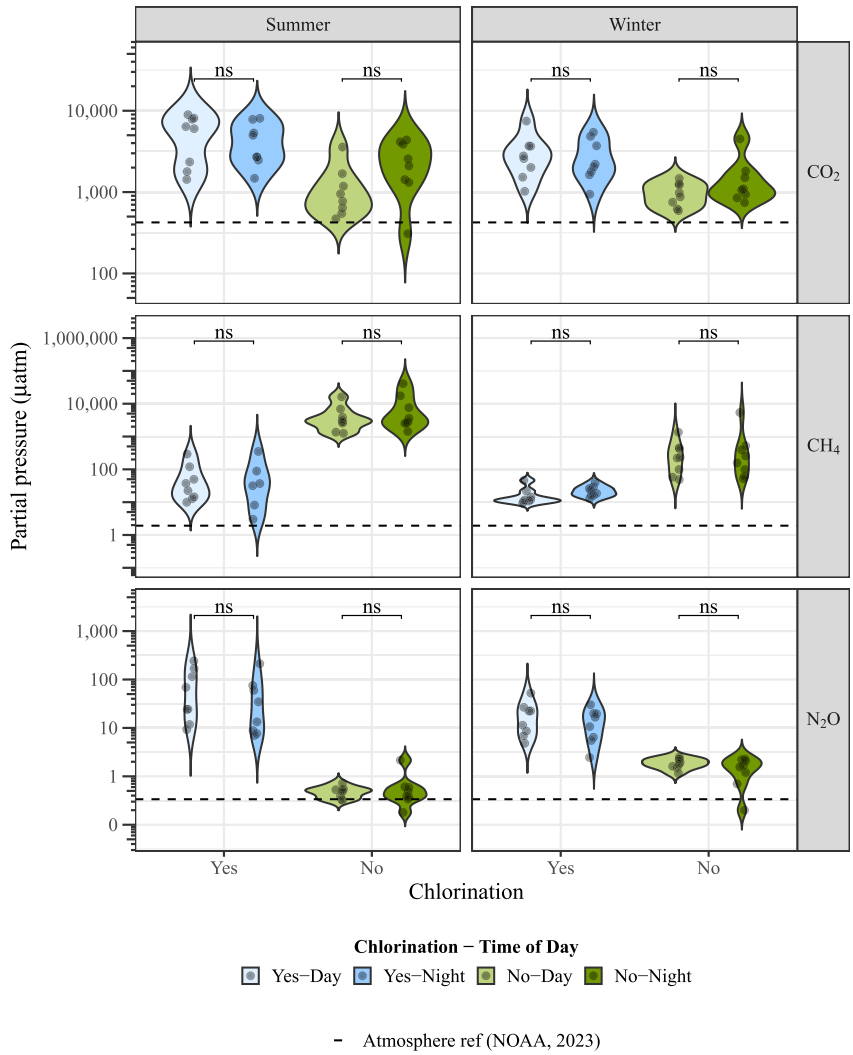

Figure 4. Density plot of partial pressure (µatm) for CO_2, CH_4 and N_2O in chlorinated (blue) and non-chlorinated (green) urban ponds measured during day (lighter shading) and night (darker shading). Panels on the left correspond to summer, and panels on the right correspond to winter. Points represent individual measurements. Pairwise comparisons between night and day measurements are indicated above the brackets, with "ns" denoting nonsignificant difference. The dashed line indicates the reference value of partial pressure in the atmosphere according to NOAA 2023 (https://gml.noaa.gov/ccgg/trends/). Note log-scale in the *y*-axis.

Table 1
Summary of the Most Parsimonious Model Selection Results for Predicting Partial Pressures of GHGs (pCO_2, pCH_4, and pN_2O) in Urban Ponds

Model	DF	RSE	R^2	R^2adj	p-value	F	AIC
$pCO_2 \sim$ Temp − DO − Fe + Mn − pH + SurfaceCover	74	0.56	0.65	0.62	***	23.10	145.31
$pCH_4 \sim$ Temp − SO_4 − DO + Chla + SubsurfaceCover	76	1.07	0.72	0.7	***	39.33	251.34
$pN_2O \sim$ −Temp−SO_4 − Fe + Mn + NO_3 − pH − SubsurfaceCover	74	0.89	0.74	0.72	***	30.77	223.09

Note. The models were selected using stepwise and trimmed methods. Significant variables selected in the models are: Temp (water temperature), Fe (dissolved iron), Mn (dissolved manganese), SO_4 (sulfate), NO_3 (nitrate), SurfaceCover (percentage of surface vegetation cover), DO (dissolved oxygen), pH, Chla (chlorophyll-*a*), and SubsurfaceCover (percentage of submerged vegetation cover). The table includes degrees of freedom (DF), residual standard error (SE), R^2, adjusted R^2 (R^2adj), *p*-value, *F*-statistic (*F*), and Akaike Information Criterion (AIC). Remaining variables are listed in Table S1 in Supporting Information S1 were also included in the analysis but were discarded during the stepwise model selection. *** indicates a *p*-value lower than 0.001.

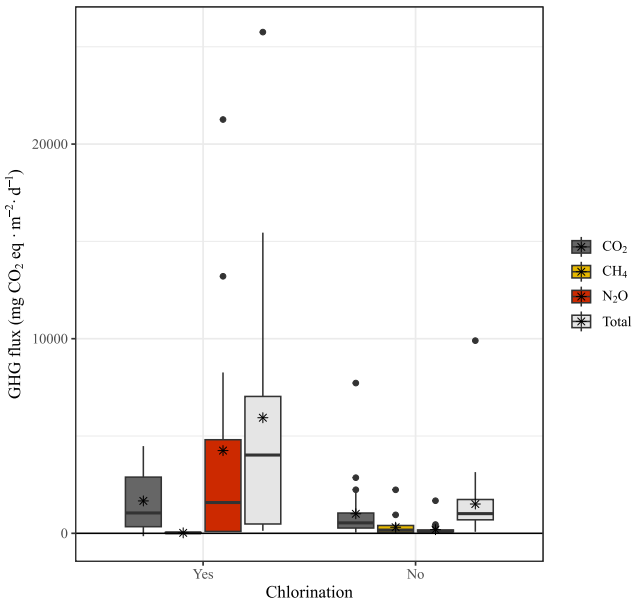

Figure 5. Estimated diffusive flux of CO_2, CH_4, and N_2O, and the total diffusive flux expressed as CO_2 equivalent (CO_2eq) in terms of global warming potential (GWP) for chlorinated (left) and non-chlorinated (right) urban ponds. GWP values of 27 and 273 times that of CO_2 for a 100-year timescale were applied for CH_4 and N_2O, respectively, following the emission metric proposed in AR6 of IPCC for a 100-year horizon (IPCC, 2023). The horizontal line within each box represents the median flux. The edges of the boxes indicate the first quartile (Q1) and the third quartile (Q3). The vertical lines (whiskers) extend to the minimum and maximum values within 1.5 times the interquartile range (IQR) from the quartiles. Asterisks represent mean values, and points show outliers.

~40% of the emissions from these ponds. Nevertheless, the CO_2 flux in chlorinated AUP was not significantly different from the flux in non-chlorinated AUP ($W = 217$, $p = 0.22$).

The CH_4 flux was significantly higher in non-chlorinated AUP [168 (60, 403) mg CO_2eq·m^{-2} · d^{-1}] than in chlorinated AUP [25 (4, 43) CO_2eq·m^{-2} · d^{-1}] ($W = 45$, $p < 0.001$). Furthermore, the contribution of CH_4 to the total flux was ~20% in non-chlorinated AUP, whereas it was lower than 1% in chlorinated AUP.

The N_2O flux was substantially higher in chlorinated AUP [1,582 (97, 4,809) mg CO_2eq·m^{-2} · d^{-1}] compared to non-chlorinated AUP [108 (78, 173) mg CO_2eq·m^{-2} · d^{-1}] ($W = 282$, $p < 0.01$). This resulted in N_2O being the highest contributor to the total flux from chlorinated AUP (~50%), whereas it was less relevant in non-chlorinated AUP (~20%).

4. Discussion

Our findings indicate that chlorination cessation does not significantly increase GHG emissions from AUP. The evidence points to a supersaturated groundwater source as a contributor to the elevated partial pressures of GHGs observed in AUP. Under these circumstances, chlorination cessation has no significant effect on pCO_2 but increases pCH_4 and reduces pN_2O, resulting in a comparable net CO_2-equivalent flux between chlorinated and non-chlorinated AUP. The effect of chlorination cessation is more pronounced in summer, likely due to increased metabolic activity. Moreover, our results demonstrate that sampling during daytime does not introduce a significant bias in GHG partial pressure measurements in AUP, possibly due to the small and shallow characteristics of these systems.

4.1. Effects of Chlorination Cessation

Most AUP in Barcelona were found to be supersaturated with respect to the atmosphere for all three studied GHGs (CO_2, CH_4, and N_2O). This situation is often reported for both natural (Holgerson & Raymond, 2016; Malyan et al., 2022) and artificial/urban ponds (Audet et al., 2020; Bauduin et al., 2024a, 2024b; Peacock et al., 2019, 2021). It is linked to the high intensity of biogeochemical processes occurring in these ecosystems (Downing, 2010), as well as the prevalence of microbial processes that favor the presence of GHGs, such as N_2O, in the groundwater used to supply the urban ponds.

The pCO_2 and pCH_4 observed in our study were consistent with previous research in urban ponds, with maximum pCO_2 values comparable to those reported by Peacock et al. (2019) and Wang et al. (2021), and pCH_4 levels falling within the range observed in studies by Bauduin et al. (2024a, 2024b); Peacock et al. (2019), and Audet et al. (2020). Although information on pN_2O is scarce, the values measured in our study were remarkably high. However, the median pN_2O in non-chlorinated AUP was lower than the average reported for other non-chlorinated/naturalized urban ponds (Audet et al., 2020; Bauduin et al., 2024a, 2024b).

Unexpectedly, chlorinated AUP were also supersaturated in CO_2, CH_4, and N_2O. The use of groundwater to refill AUP in the city of Barcelona is a common practice, and groundwater is usually supersaturated in these gases (Jahangir et al., 2012; Jurado et al., 2017; Nikolenko et al., 2019). In fact, the partial pressures measured at 7 groundwater extraction points in the city of Barcelona were 13,909 [11,073, 18,058] µatm of CO_2, 12.53 [4.84, 18.06] µatm of CH_4, and 19.06 [11.83, 41.63] µatm of N_2O. If this supersaturated groundwater was used to refill AUP, the partial pressures measured in chlorinated AUP, where biological activity is expected to be minimal, would be lower due to gas loss from degassing and equilibration with the atmosphere. Indeed, our findings support this: partial pressures in chlorinated AUP were supersaturated but slightly lower than those in the groundwater, supporting the hypothesis of a possible groundwater origin for the GHGs measured in these AUP.

The pCO_2 was similar in both chlorinated and non-chlorinated AUP. This result can be attributed to the high environmental variability within non-chlorinated AUP, particularly in terms of vegetation cover and chlorophyll-

a concentration (Table S1 in Supporting Information S1). The intrinsic variability of the AUP may result in ecosystems ranging from autotrophic (net ecosystem production > 0) to heterotrophic (net ecosystem production < 0). Moreover, individual ponds may shift between autotrophic and heterotrophic states depending on the season (Bauduin et al., 2024a, 2024b; Rackliffe et al., 2021).

Non-chlorinated AUP showed significantly higher pCH_4 than chlorinated AUP, falling within the range typically found in inland waters (Malerba et al., 2022; Rocher-Ros et al., 2023) and comparable to those found in other non-chlorinated/naturalized urban ponds (Audet et al., 2020; Bauduin et al., 2024a, 2024b; Peacock et al., 2021). This elevated pCH_4 can be attributed to enhanced methanogenesis in these naturalized ecosystems, whereby CH_4 is mainly produced by microorganism under anaerobic conditions (Bastviken, 2009). Conversely, in AUP under chlorine treatment, absence of anaerobic conditions and negligible microbial activity are expected.

In contrast with pCH_4, we found higher pN_2O in chlorinated AUP than in non-chlorinated AUP. The pN_2O measured in chlorinated AUP felt within the range reported for groundwater, where N_2O is known to reach much higher concentrations than in surface waters (Jahangir et al., 2012; Jurado et al., 2017). In fact, most of our recorded values were supersaturated but below the mean concentrations typically reported for groundwater (Jurado et al., 2017). The groundwater of Barcelona presents favorable conditions for the denitrification process, which makes it an initial source of N_2O for the medium. We hypothesize that the lower pN_2O found in non-chlorinated AUP may be due to microbial activity (i.e., denitrification), which can consume N_2O (Pan et al., 2023; Pauleta et al., 2019) and potentially act as an effective N_2O sink (Conthe et al., 2019). When N_2O-rich groundwater enters non-chlorinated AUP, which present methanogenic conditions evidenced by elevated CH_4 concentrations, N_2O may be reduced via denitrification. This results in a higher consumption of N_2O and its conversion to N_2 (Espenberg et al., 2024) in non-chlorinated AUP compared to chlorinated AUP. Our findings highlight that non-chlorinated ponds act as effective N_2O sinks, contributing to the mitigation of this GHG in the environment (Conthe et al., 2019).

4.2. Temporal Variability: Seasonal and Daily Changes

In non-chlorinated AUP, we observed seasonal variation in pCH_4 and pN_2O but not in pCO_2. In contrast, no significant seasonal differences in GHG partial pressures were observed in chlorinated AUP.

The differences in pCH_4 and pN_2O observed in non-chlorinated AUP are likely linked to biological activity. In summer, higher temperatures favor metabolic processes, leading to increased methanogenesis and, consequently, higher CH_4 concentrations (Yvon-Durocher et al., 2014). Similarly, denitrification is a temperature sensitive process (Velthuis & Veraart, 2022) and, therefore, denitrification is expected to be higher in summer. This higher activity can reduce N_2O to N_2 (Pan et al., 2023; Pauleta et al., 2019) thereby diminishing pN_2O. The absence of seasonal differences in pCO_2 contrasts with findings by Wang et al. (2023), who attribute seasonal variability in urban freshwater ecosystems to two main processes: strong internal respiration under higher summer temperatures and increased microbial activity in surrounding soils. However, microbial activity in surrounding soils is unlikely to play a significant role in our case, as the studied AUPs are surrounded by concrete, and little to no subsurface water from surrounding soils is expected to enter the ponds. Regarding internal respiration, in our AUPs, this process may be masked by the activity of primary producers. Although Wang et al. (2023) focused on urban rivers and lakes, our study focused on urban ponds, which typically present a higher proportion of primary producers. In fact, we observed surface and subsurface vegetation cover reaching up to 100%. This higher abundance of primary producers may buffer the effect of microbial respiration, minimizing seasonal variability in pCO_2.

No significant differences were found between day and night measurements for any of the measured GHGs (CO_2, CH_4, and N_2O) in chlorinated and non-chlorinated AUP. In the chlorinated AUP, we did not expect differences between day and night due to their limited biological activity. However, it was surprising not to find significant diel changes in the non-chlorinated AUP. A potential explanation for the lack of day-night differences in GHG partial pressures may be related to pronounced physical stratification in the study ponds. Holgerson et al. (2022) showed that small ponds deeper than 0.74 m rarely or never mix due to limited wind energy relative to their density gradient. Non-chlorinated AUP sampled at night were deeper than 0.7 m and often surrounded by buildings and other infrastructures that protected them from wind. Stratification could have prevented mixing with deeper layers, where respiration by submerged macrophytes can lead to anoxia, and where methanogenesis and denitrification can be more intense (Rehder et al., 2021). Consequently, GHG partial pressures below the

water surface likely changed slowly through molecular diffusion and therefore did not experience significant differences between day and night.

4.3. Drivers

Our results indicate that naturalization, groundwater legacy, and season are the main drivers influencing GHG partial pressures in AUP.

Percentage of surface and submerged vegetation cover, dissolved oxygen and NO_3^- are variables related to naturalization. Submerged and surface vegetation influence GHG dynamics through different mechanisms. Surface vegetation cover was associated with higher pCO_2 in the studied AUP, possibly due to the shading effect of the vegetation, which suppresses primary production and enhances respiration (Rackliffe et al., 2021). Submerged vegetation cover showed a positive relationship with CH_4, a finding consistent with previous studies in urban ponds (Hilt et al., 2022; Natchimuthu et al., 2014). This relationship may be attributed to the establishment of anoxic conditions and the contribution of organic matter, which enhance methanogenesis (Hilt et al., 2022; Theus et al., 2023; Vilas et al., 2017). Similarly, chlorophyll-*a* has been positively linked to CH_4 production (Beaulieu et al., 2019; Colas et al., 2021; DelSontro et al., 2018), suggesting that elevated primary production may influence CH_4 dynamics. Dissolved oxygen is related to both primary production and respiration and, therefore, expected to show a direct relationship with pCO_2. High rates of aerobic respiration, coupled with low primary production, can lead to increased pCO_2. This imbalance may result in anaerobic conditions, which can favor CH_4 production. NO_3^- was positively related with pN_2O. This could be explained by denitrification, which not only consumes NO_3^- but can also serve as a sink for N_2O (Conthe et al., 2019).

The relationships between Fe and Mn with CO_2 and N_2O, as well as SO_4^{2-} with N_2O, may be linked to groundwater legacy. In Barcelona, groundwater is widely used to maintain pond water levels. Urban groundwater often experiences low dissolved oxygen conditions (Datry et al., 2004; Koch et al., 2021). These conditions promote N_2O production through nitrification and denitrification processes (Jurado et al., 2017; Nikolenko et al., 2019) while also leading to the accumulation of CO_2 produced via respiration processes (Wood & Petraitis, 1984). Additionally, hypoxic waters also favor sulfate reduction, resulting in SO_4^{2-} depletion. Lastly, Fe and Mn are elements involved in anaerobic metabolism (Stumm & Morgan, 1996) and in the abiotic production of N_2O through chemodenitrification and chemical oxidation of hydroxylamine (Pan et al., 2023).

Temperature is directly related to season and is a well-established factor in regulating metabolic processes in aquatic ecosystems (Brown et al., 2004). Linear mixed models revealed significant relationships between temperature and all three GHGs (CO_2, CH_4, and N_2O). Specifically, higher temperatures were positively related with pCO_2 and pCH_4, likely indicating enhanced aerobic respiration and methanogenesis under higher temperature (Malyan et al., 2022; Marotta et al., 2014). In contrast, N_2O showed a negative relation with temperature. Higher temperatures are associated with increased denitrification activity (Velthuis & Veraart, 2022), which can reduce N_2O from groundwater sources to N_2 (Pan et al., 2023; Pauleta et al., 2019), thereby, lowering pN_2O.

4.4. Diffusive Fluxes

Both chlorinated and non-chlorinated AUP of Barcelona city were net emitters of CO_2, CH_4, and N_2O. Overall, both pond types showed a similar flux in CO_2eq. However, the contribution of each gas was different depending on whether the AUP were chlorinated or non-chlorinated.

Non-chlorinated AUP emitted more CH_4, mainly due to the abundance of primary producers (macrophytes and phytoplankton), which create suitable conditions for microbial activity and, consequently, methanogenesis. Moreover, our results indicated that chlorination cessation can also enhance denitrification, which can help reduce the pN_2O when water input is supersaturated with N_2O. Regarding CO_2, no significant differences were found between pond types, likely due to the high variability between autotrophic and heterotrophic net ecosystem metabolism in non-chlorinated AUP.

Emissions from chlorinated AUP are determined by the source of the water. Although the origin of the water might appear insignificant, when GHG supersaturated groundwater is used to refill the AUP, they can act as emitters through degassing, similar to what has been reported for ponds subject to agricultural irrigation (Huo et al., 2022; Huo & Gao, 2024; McGill et al., 2018).

Estimated emissions were within the range reported for CO_2 (Goedker et al., 2022; Peacock et al., 2021; Wang et al., 2021) and CH_4 (Audet et al., 2020; Gorsky et al., 2019; van Bergen et al., 2019) in urban ponds. However, N_2O emissions were higher than those reported for urban ponds (Bauduin et al., 2024a, 2024b; Ray et al., 2023) but similar to the degassing flux from groundwater irrigation found by Huo and Gao (2024).

It is important to emphasize that these emissions are negligible when put into the context of a city such as Barcelona, which is located in a semi-arid, water-scarce region. The city of Barcelona has a surface area of artificial water bodies covering approximately 13 ha (data provided by the Barcelona Council), which according to our data results in a rough estimate of annual emissions of approximately 57 [29, 114] $t\,CO_2eq{\cdot}year^{-1}$. The total annual emissions for the city amount to 3,557,530 t CO_2eq, with an annual emission per inhabitant of 2.17 t CO_2eq (Agència d'energia de Barcelona, 2019). Emissions from AUP would therefore represent less than 0.002% of the total CO_2eq emissions for the city, equivalent to the annual emissions of 27 residents. Given the significant benefits that these blue ecosystems can provide to society, such as increased biodiversity, water purification, and enhanced human well-being (Oertli et al., 2023), this represents a minimal ecological footprint. It is important to note that the significance of AUP and other urban water bodies may vary in cities located in more humid regions, where these ecosystems tend to cover a considerably larger area.

4.5. Limitations and Future Research

In this study, we measured GHG partial pressures in the water of AUP. We showed that the chlorination cessation in AUP affects pCH_4 and pN_2O but not pCO_2. Most biogeochemical processes in these ecosystems interact directly with the water column, and GHG exchange with the atmosphere occurs mainly at the water surface. Here, the uncertainty related to the estimation of k_{600} can play a relevant role (see Section 2.4). Moreover, emergent and floating vegetation also exchange GHGs directly with the atmosphere. This plant-atmosphere exchange can significantly influence the net balance by increasing CH_4 emissions through aerenchyma or by consuming/emitting CO_2 (Attermeyer et al., 2016). In addition, ebullition in urban inland waters can represent an important source of CH_4 (Wang et al., 2021). Therefore, future studies should include direct measurements of GHG fluxes from exposed plant tissues as well as ebullition to obtain a comprehensive understanding and establish a more integrated view of the effect of naturalization on the GHG balance of urban ponds. Furthermore, we demonstrated that AUP can be a source of GHGs. However, natural ponds can also show high carbon burial rates (Taylor et al., 2019). Despite this, little is known about the carbon burial capacity of AUP. Investigating their carbon sequestration potential would provide a deeper understanding of their role in climate regulation services.

Data Availability Statement

The data used in the study are available at: https://portal.edirepository.org/nis/mapbrowse?packageid=edi.1927.2 (Montes-Pérez et al., 2025).

The code used to reproduce all results and figures presented in this article is available at: https://doi.org/10.5281/zenodo.16628631 (Montes-Pérez, 2025).

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