Occurrence and flux of polycyclic aromatic hydrocarbons in settling particulate matter from the water column in Fildes Bay, Antarctica

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Abstract

This study investigates the occurrence, sources and fluxes of polycyclic aromatic hydrocarbons (PAHs) in settling particulate matter (SPM) from the water column from two locations in Fildes Bay (King George Island), Antarctica. Fildes Bay is an area influenced by research stations, maritime activities, and tourism. The SPM sample was obtained from a Sediment trap (20–50 m depth) during summer Antarctic expeditions. In this work, which complements our previous study regarding of PAHs (18), PAHs were quantified by gas chromatography–mass spectrometry (GC–MS). The results show Σ18PAHs ranged from 2 to 18 ng g^{-1} dry weight (DW), in which PAHs predominate 2- to 3- ring. The fluxes from SPM in the water column ranged from 0.03 to 0.70 ng m^{-2} day^{-1} and increased near Collins Glacier. The temporal variability observed during the sampling period highlights the influence of both natural and anthropogenic factors. These findings contribute to the scarce dataset available for Antarctic marine environments and emphasize the importance of long-term monitoring to assess pollutant dynamics under changing climatic conditions.

Introduction

Antarctica is often seen as one of the last untouched environments on Earth (Vodopivez et al. 2021). However, increasing evidence indicates that this remote continent is gradually impacted by human-caused pollution (Bargagli 2008). Several studies have documented the presence of contaminants such as heavy metals (Chu et al. 2019, Chiang et al. 2021), persistent organic pollutants (POPs) (Nash 2011, Vecchiato et al. 2025, Luarte et al. 2023), and polycyclic aromatic hydrocarbons (PAHs) (Cabrerizo et al. 2014, Szopińska et al. 2019, Wu et al. 2013) in various Antarctic environmental matrices.

PAHs are of particular concern due to their toxicity, persistence, and bioaccumulative potential in polar ecosystems (Cabrerizo et al. 2014, Almeda et al. 2021, Xie et al. 2022). These compounds mainly originate from the incomplete combustion of organic matter, including fossil fuels, biomass burning, and industrial emissions (Stogiannidis and Laane 2015, Pozo et al. 2023). In remote areas like Antarctica, PAHs can be introduced through long-range atmospheric transport and deposited via wet or dry atmospheric deposition, often adsorbed onto fine suspended particles. Maritime activities (marine operation of ships and vessels) and local human presence also contribute to their input due to energy generation activities in local settlements (Chiuchiolo et al. 2004, Na et al. 2011, Cabrerizo et al. 2014).

Polycyclic aromatic hydrocarbons (PAHs) are semi-volatile, hydrophobic organic pollutants that tend to associate with particulate matter, especially those rich in organic carbon or lipids, leading to their accumulation in marine sediments (Baran et al. 2017). In polar environments, these compounds can enter seawater, sea ice, sediments, and biota (Curtosi et al. 2009), where their fate is determined by complex interactions between partitioning processes, degradation rates–including microbial degradation–and vertical transport mechanisms (Duran and Cravo-Laureau 2016). Vertical transport is closely connected to the biological pump–a process by which organic matter produced in the photic zone, often through phytoplankton activity, aggregates and sinks, carrying associated contaminants such as PAHs (Dang and Lovell 2016). The efficiency of this process is influenced by seasonal biological productivity, ice dynamics, and turbulence in the water column (Ding et al. 2021). Additionally, degradation processes, both photo-

chemical and microbial, may alter the concentration and composition of PAHs during their transport, as demonstrated in other regions of the world (Gonzalez-Gaya et al. 2019).

Suspended particulate matter acts as a major carrier for polycyclic aromatic hydrocarbons (PAHs) in the water column, aiding their downward movement from surface waters to deeper layers. These particles not only influence the vertical distribution and environmental fate of PAHs but also function as temporary reservoirs. Under changing hydrodynamic conditions–such as turbulence, density gradients, or biological activity–PAHs can be remobilized through processes like desorption or resuspension, potentially increasing their bioavailability or redistributing them within the column (González-Gaya 2015, Loh et al. 2018).

In this context, Sediment traps serve as powerful tools for investigating the fate of particle-bound PAHs (Lipiatou et al. 1993, Skylaki et al. 2025). By capturing sinking particulate matter over time, they allow for the quantification of vertical settling fluxes and the associated contaminant loads. Providing critical insights into the pollutant biogeochemical cycles and in polar systems where physical and biological processes are coupled.

Fildes Bay, located on King George Island in the South Shetland Islands, is a semi-enclosed coastal system subject to both natural polar dynamics and increasing anthropogenic influence due to the presence of multiple scientific research stations and associated human activity (Potapowicz et al. 2022, Wu et al. 2023). Its relatively shallow depth, seasonal ice cover, and variable hydrodynamic conditions make it a particularly dynamic environment for the study of contaminant behaviour. In this context, Fildes Bay is a sensitive site for evaluating the occurrence and flux of polycyclic aromatic hydrocarbons (PAHs) associated with sedimentation particles, due to the aforementioned human influence activities. The bay's seasonal biological productivity, coupled with episodic resuspension events and ice melt, creates favourable conditions for the vertical transport of particle-bound contaminants. Investigating PAH concentrations and fluxes in the water column of Fildes Bay offers valuable insights into the environmental fate of these pollutants in Antarctic coastal systems and contributes to the broader understanding of pollutant cycling in rapidly changing polar environments.

This study aims to assess the occurrence, characterize the profile of PAHs and sources associated with settling particulate matter collected using Sediment traps at two sampling sites in Fildes Bay. Through this analysis, we aim to contribute to the understanding of contaminant transport and accumulation processes in polar marine ecosystems, providing insights into their distribution and potential ecological impacts.

Material and Methods

Sampling and study area

Sampling was carried out at two different sites at Fildes Bay (also known as Maxwell Bay), on King George Island, South Shetland Archipelago, Antarctica (Sediment trap 1 (62°11′10″ S, 58°48′ 50″ W) and Sediment trap 2 (62°12′15″ S, 58°56′35″ W)) (*see* Fig. 1a) from December 16th, 2019, to January 27th, 2020 (Table 1). For sampling, a Sediment trap sampling was used (*see* Fig. 1b). The Sediment trap was equipped with 9 x 40 cm acrylic tubes (4 tubes per trap), each tube had a PVC valve at the bottom for sample output. The traps were installed with a moor-

ing system reaching approximately 50 m of the water column. Sampling was carried out in parallel at each site and lasted 4 to 6 days. The samples were collected in previously cleaned and calcined glass jars on board the vessel. Samples were processed immediately upon arrival at the Professor Julio Escudero Scientific Base laboratory, within two hours of collection, to ensure preservation of sample integrity before chemical analysis.

Once in the laboratory, the sample collected were filtered using GF/F filters (47 mm diameter, 0.7 µm pore size, Whatman™) using a vacuum filtration system. Samples collected were kept -20°C and freeze-dried until their subsequent extraction and analysis.

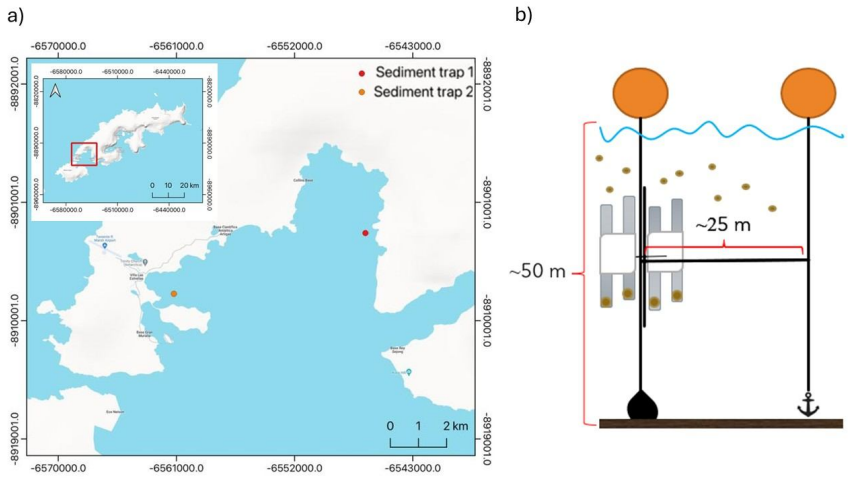

Fig. 1. a) Study area and b) Sediment trap deployment scheme in Fildes Bay, Antarctica.

Start date	End date	Days	Sample site 1	Sample site 2
16-12-2019	21-12-2019	5	Sediment trap1_1	Sediment trap 2_1
21-12-2019	27-12-2019	6	Sediment trap1_2	Sediment trap 2_2
27-12-2019	03-01-2020	6	Sediment trap1_3	Sediment trap 2_3
03-01-2020	08-01-2020	5	Sediment trap1_4	Sediment trap 2_4
08-01-2020	13-01-2020	5	Sediment trap1_5	Sediment trap 2_5
13-01-2020	17-01-2029	4	Sediment trap1_6	Sediment trap 2_6
17-01-2020	21-01-2020	4	Sediment trap1_7	Sediment trap 2_7
21-01-2020	27-01-2020	6	Sediment trap1_8	Sediment trap 2_8

Table 1. Sampling periods and corresponding Sediment trap deployments at sites 1 and 2 (December 2019 – January 2020).

Chemical analysis

Samples were spiked with PAHs surrogate standards (Naphthalene-D8, Acenaphthene-D10, Phenanthrene-D10, Pyrene -D10, Benzo(a)anthracene-D12, Perylene-D12 (250 ng/sample))(Sigma-Aldrich, MA, USA) before chemical analysis. Subsequently, the samples were Soxhlet-extracted using 150 mL of dichloromethane in an automated extractor system (Büchi B-811, Switzerland) with the program set to hot Soxhlet extraction for 40 minutes and washing with solvent condensate for 20 minutes.

Samples were cleaned using a non-destructive silica glass column (1 cm i.d.), containing 5 g of activated silica and 1 cm of anhydrous Na_2SO_4, eluted with 10 mL of *n*-hexane and 40 mL of dichloromethane. Samples were concentrated to 1 mL using SuperVap (FMS, MA, USA) and transferred to 1,5 mL GC vials. 50 µL of nonane was added as a keeper solvent, and samples were concentrated under a gentle stream of nitrogen in LabEva to a final volume of 50 µL. Before instrumental analysis, samples were spiked with *p*-terphenyl (200 ng), Wellington, MA, USA) for determination of % recoveries of surrogate (internal) standards.

Polycyclic aromatic hydrocarbons (PAHs) were analyzed into individual species using an 8890A GC (Agilent, USA) equipped with a 30 m × 0.25 mm × 0.20 µm ZB-PAHs-CT column (Phenomenex, USA), coupled to a triple quadrupole 7000D MS (Agilent, USA). The GC oven temperature program started at 80°C (2 min hold), then increased at 15°C/min. to 180°C (no hold), and finally at 5°C/min. to 310°C (20 min. hold). The inlet temperature was set to 280°C, with an injection volume of 1 µL in pulsed-splitless mode. Helium served as the carrier gas at a flow rate of 1.5 mL/min. The GC-MS transfer line temperature was 310°C, and the ion source was heated to 320°C. The mass spectrometer operated in multiple reaction monitoring (MRM) mode, with nitrogen as the carrier gas at 1.5 mL/min. Compound quantification was performed with MassHunter Workstation 10.1 software. The following PAHs were assessed: acenaphthene (ACE), fluorene (FLN), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benz(a)anthracene (BAA), chrysene (CHR), benzo(b)fluoranthene (BBF), benzo(a)pyrene (BAP), indeno(1,2,3-cd)pyrene (INP), benzo(g,h,i)perylene (BPE), biphenyl (BIF), benzo(b)fluorene (BBFL), benzo(naphtho) thiophene (BNT), benzo(g,h,i)fluoranthene (BGF), triphenylene (TRI), and benzo(e) pyrene (BEP).

Quality assurance/quality control (QA/QC)

The analytical procedures described above were verified for recoveries and reproducibility. Method recoveries for PAHs were evaluated based on recoveries of three deuterated PAHs added prior to sample preparation: Naphthalene-d8 (60.8% ± 15.9), phenanthrene-d10 (70.6% ± 21.8), and perylene-d12 (94.5% ± 4.1). Blank levels were measured using field blanks (n = 2) and laboratory blanks (n = 2 solvent blanks). The instrumental detection limits (IDL) were determined by assessing the injection amount that produced a signal-to-noise ratio ≥ 3. Method detection limits (MDL) in the samples were calculated as the average of the combined field and laboratory blanks (n = 4) plus standard deviations (SD). When target compounds were detected (< 10%) in blanks, half of the IDL value was used. All PAHs were quantified using external calibration over a linear range from 1 to 1000 ng/sample. To estimate the actual concentration (ng g^{-1}), multiply the result by the weight of the filters (initial weight minus final weight).

PAH sources using diagnostic ratios

The common diagnostic ratios widely applied in the literature consisting of ANT/(ANT + PHE), BAA/(BAA + CHR), FLT/(FLT + PYR), and INP/(INP + BPE), were employed in this study to evaluate the potential sources of PAHs (Luo et al. 2019). An ANT/(ANT + PHE) ratio < 0.1 implies petroleum contamination, while > 0.1 means that the components originate from combustion (Xue et al. 2014). A BAA/(BAA + CHR) ratio > 0.35 indicates vehicle emission sources, < 0.2 suggests oil pollution, while 0.2–0.35 indicates mixed oil and diesel combustion sources (Sarigiannis et al. 2017). A FLT/(FLT + PYR) ratio < 0.4 indicates oil volatilization, > 0.5 indicates coal and biomass combustion, while 0.4–0.5 indicates oil burning (Omar et al. 2006). An INP/(INP + BPE) isomeric ratio < 0.2 represents oil combustion, > 0.5 indicates partial combustion of coal, wood, or grass, and 0.2–0.5 represents fossil fuel combustion (Jamhari et al. 2014).

Statistical analysis

The statistical analysis was conducted using GraphPad Prism software (version 10.4.2). A principal component analysis (PCA) was first performed to investigate the grouping patterns of polycyclic aromatic hydrocarbons (PAHs), their number of aromatic rings and relative abundance. The data were normalized, and principal components were calculated to generate a biplot. Additionally, boxplot analyses were carried out to assess the distribution and trends of diagnostic ratios commonly used to identify potential sources of PAHs, with comparisons made between the different sampling locations. Finally, a line graph was constructed to compare the vertical fluxes of PAHs at the two study sites, providing insights into the temporal and spatial variability of these compounds in the samples.

Results and Discussion

PAH levels and fingerprints

The average concentrations of the 18 polycyclic aromatic hydrocarbons (Σ18PAHs) showed a clear difference between the two sampling sites, with Sediment trap 1 reporting 11.4 ± 11.5 ng g^{-1} DW and Sediment trap 2 reporting 4.2 ± 3.1 ng g^{-1} DW. Overall, Sediment trap 1 exhibited higher variability for most compounds (Fig. 2), especially Fluorene (FLN), Phenanthrene (PHE), Anthracene (ANT), and Benzo(g,h,i)perylene (BPE), whose levels were significantly higher than those recorded at Sediment trap 2. This trend was consistent in both absolute values and relative composition, as shown in Fig. 2a. The apparent higher abundance observed at Sediment trap 1 compared to Sediment trap 2 did not show any statistical difference ($p>0.05$) in terms of total PAHs. However, when comparing the abundance reported in Sediment trap 1 to that in Sediment trap 2, there is a possible influence of glacier melting during summer, which aligns with previous studies reporting higher concentrations of POPs in the bay during ice-melting periods (Austral summer). This melting could also affect the air-water exchange of Polychlorinated Biphenyls (PCBs) in the study area during the same period (Luarte et al. 2024). In contrast,

Sediment trap 2 showed lower concentrations, a more homogeneous distribution, and less influence from ice melting and particle release from the glacier. Previous studies indicating the impact of particle release near glaciers show that particle release is significant at locations close to the glacier (Syvitski 1989), as demonstrated for metals and nutrients (Fegel et al. 2016).

There are no studies on PAHs levels in settling particulate matter from the water column in Antarctica. However, a study conducted in Potter Cove, South Shetland Islands, Antarctica, reported suspended particulate matter levels in the water column ranging from 30 to 82 ng g^{-1} DW (Curtosi et al. 2009). Another study reported concentrations as high as 427 ng g^{-1} DW near Jubany Station (Curtosi et al. 2008). In our study, Σ18PAHs ranged from 2 to 18 ng g^{-1} DW, indicating that Fildes Bay does not have high PAH concentrations.

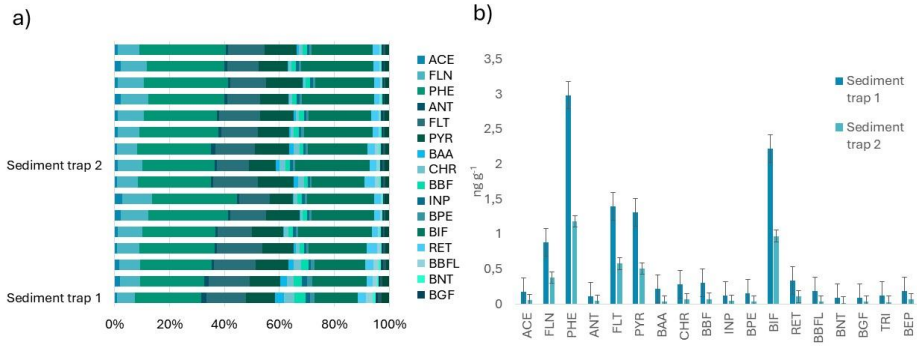

Fig. 2. Distribution and relative abundance of PAHs in suspended particulate matter from Fildes Bay, Antarctica. a) Relative composition (%) of polycyclic aromatic hydrocarbons (PAHs) in the 8 samples per site (Sediment trap 1 and Sediment trap 2), and b) Average concentration (ng g^{-1} DW) for both sites.

On the other hand, the abundance of polycyclic aromatic hydrocarbons (PAHs) by ring number was analyzed using principal component analysis (Fig. 3). When grouping PAHs by their number of rings, it was observed that 2- to 3-ring compounds were more abundant, particularly at Sediment trap 1, which is closer to the Collins Glacier, compared to Sediment trap 2, located nearer to the research station. Sediment trap 2 also shows a higher relative presence of PAHs with 4- to 6-rings. These compounds, such as ACE, FLN, and PHE, are more volatile and soluble, helping their atmospheric transport and deposition in remote areas. Four-ring PAHs, like FLT and PYR, also contributed significantly but at moderate levels, while 5- to 6-ring compounds were not detected in substantial amounts (Fig. 3). This indicates a dominance of pyrogenic sources from incomplete combustion, possibly linked to local human activity or the long-range transport of pollutants through the atmosphere. Winds may carry these contaminants from nearby sources; considering that waste incineration and energy production occur near the bases, the prevailing winds (SE, *see* Fig. 4) might meet the Collins Glacier (*see* Fig. 1), causing the deposition of pollutants near Sediment trap 1. The lower presence of high molecular weight PAHs could be due to their low volatility and limited capacity for atmospheric transport to polar regions.

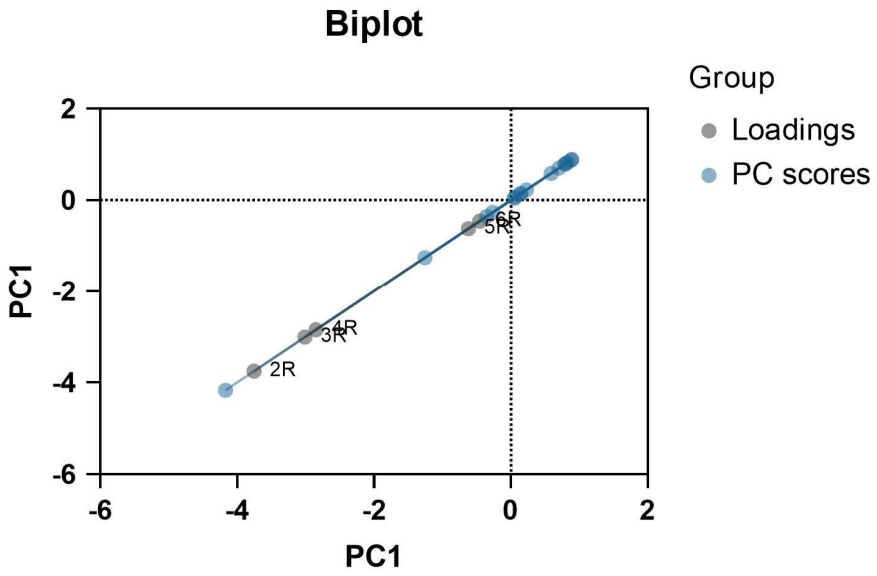

Fig. 3. Biplot of principal component analysis grouped by PAH ring number in suspended particulate matter from Fildes Bay, Antarctica.

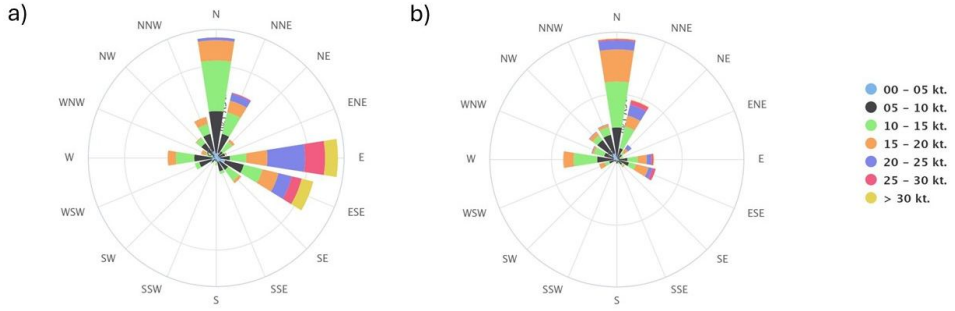

Fig. 4. Wind rose comparison for a) December 2019[1] and b) January 2020[2].

PAH sources using diagnostic ratios

Analysis of PAH diagnostic ratios reveals differences in potential sources between the two sampling sites (Table 2). At both locations, the ANT/(ANT + PHE) ratio consistently stayed below 0.1, indicating a mainly petrogenic origin for low molecular weight PAHs. However, the other ratios point to a significant contribution from pyrogenic sources. The BAA/(BA + CHR) ratio remained under 0.35 at both sites but showed more variability at Sediment trap 1, which may indicate a mixture of emissions or fluctuating combustion sources over time. For FLT/(FLT + PYR), both sites exhibited values above 0.5, suggesting a strong influence from biomass or coal combustion. Lastly, the IP/(IP + BghiP) ratio differed notably between

sites: higher and more consistent at Sediment trap 2 (>0.5), while Sediment trap 1 displayed more variable values, possibly reflecting combined effects from vehicular emissions and incomplete combustion. Overall, these findings suggest that although both sites are affected by pyrogenic sources (Fig. 5), Sediment trap 1 shows greater source diversity, likely due to local conditions or more dynamic atmospheric processes.

Site	ANT/(ANT + PHE)	BAA/(BAA + CHR)	FLT/(FLT + PYR)	INP/(INP + BPE)
Sediment trap 1	<0.1	0.2–0.35	>0.5	0.2–0.5
Sediment trap 2	<0.1	0.2–0.35	>0.5	>0.5

Table 2. Diagnostic ratios of PAHs for source identification at both sampling sites from Fildes Bay, Antarctica.

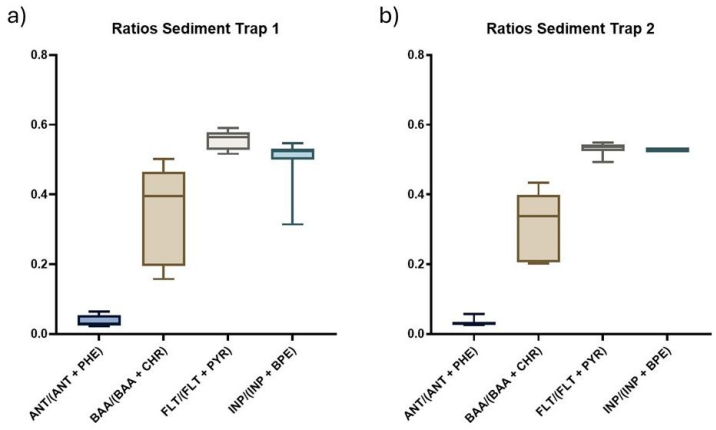

Fig. 5. Boxplots of PAH diagnostic ratios.

Fluxes

Vertical fluxes of 18 polycyclic aromatic hydrocarbons (PAHs) were calculated from 8 samples collected at each of the two sampling sites (Sediment trap 1 and 2) in Fildes Bay, Antarctica. The fluxes were estimated in ng m^{-2} day^{-1} based on the concentration of each compound (ng g^{-1}), the mass of collected particulate material (g), the effective area of the collector (0.25 m^2), and the exposure time (days) (Eqn. 1).

$$Flux = \frac{PAHs\ Concentracion\ (ng\ g^{-1}) \times SPM\ weight\ (g)}{Trap\ area\ (m^2) \times Time\ (day)} \qquad Eqn.\ 1$$

Fig. 6 shows a comparison of the average fluxes between both sites for each compound. Overall, the fluxes were consistently higher at the Sediment trap 1 site than at Sediment trap 2. The compounds with the highest average fluxes were PHE, FLT, PYR, and FLN with PHE standing out due to its significantly higher value at Sediment trap 1 (~0.75 ng m^{-2} day^{-1}) compared to Sediment trap 2 (~0.13 ng m^{-2} day^{-1}).

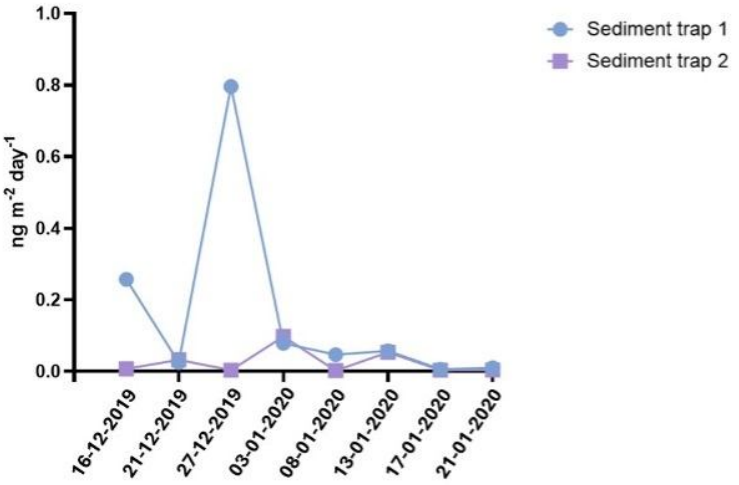

Fig. 6. Comparison of vertical fluxes of PAHs (ng m^{-2} day^{-1}) per sample at two Sediment trap sites in Fildes Bay, Antactica.

The differences observed in PAH fluxes between Sediment trap 1 and 2 may be linked to various environmental and human-related factors. The line graph highlights a significant difference in the magnitude and variability of PAH fluxes between the two sites. Sediment trap 1 exhibits a clear peak in sample 3, with a flux exceeding 0.80 ng m^{-2} day^{-1} (Fig. 6), while the other samples at that site show much lower values. This peak indicates a potential local release of contaminants or specific weather conditions that favored deposition. Conversely, Sediment trap 2 displays more consistent and lower fluxes across all samples, with a peak in sample 4 (~0.1 ng m^{-2} day^{-1}) (Fig. 6), without sudden changes.

Sediment trap 1 may receive a similar atmospheric load of pollutants compared to Sediment trap 2, but it is also influenced by melting from the glacier, as these compounds could accumulate over time and be released during melting periods (Casla et al. 2018, Zenteno et al. 2019, Luarte et al. 2025). Additionally, this area is affected by nutrient-rich waters from glacial melt. Luarte et al. (2025) reported elevated chlorophyll concentrations in Fildes Bay during the same period (especially on day 3) when samples were collected, coinciding with the sampling dates that showed the highest pollutant fluxes. Furthermore, the influence of ice melting has been shown to have a greater impact on PAH levels in surface waters in areas highly affected by

melting (low salinity and high temperature), as noted in other Antarctic coastal regions (Casal et al. 2018). This significant influence may alter sedimentation rates and the levels of PAHs in marine particulate matter. Since low temperatures can affect the partitioning of dissolved PAHs in the water column, shifting the equilibrium from dissolved to particulate matter, and this, combined with a possible increase in dissolved PAHs due to ice melting, may influence particulate PAH levels (Lüers and Ten Huschler 1996). Consequently, particulate matter affects PAH concentrations and the subsequent sedimentation rates.

Conclusion

Although Antarctica is still one of the least polluted areas on Earth, the detection of PAHs in SPM underscores the widespread influence of human activities and their long-term effects on even the most remote regions. Due to the persistence and ecological dangers posed by these contaminants, ongoing monitoring, stricter regulations, and better waste management at research stations and tourism sites are crucial to safeguarding Antarctic ecosystems. Future research should explore emerging pollutants, how climate change alters pollutant behaviour, and how international regulations are helping to lower PAH levels in the region.

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