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Key Points:
- Polycyclic aromatic hydrocarbons (PAHs) generally exhibited a surface enrichment and depth depletion pattern throughout the water in the Fram Strait
- PAHs are released from the Arctic Ocean to the North Atlantic through the deep-water passage Fram Strait and surface pathway Davis Strait
- PAHs in the Arctic and Great Lake regions were mainly derived from pyrogenic sources, with the existence of some biomass burning sources

Supporting Information:

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

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Presence, Sources and Transport of Polycyclic Aromatic Hydrocarbons in the Arctic Ocean

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Abstract Polycyclic aromatic hydrocarbons (PAHs) are continuously released from multiple sources and are prone to long-range transport. Little is known regarding their presence, transport and fate in remote and deep oceans. Polyethylene passive samplers were hence deployed at deep moorings and surface seawater in the Fram Strait and Canadian Archipelago, as well as in air and surface water of the lower Great Lakes, a potential high-emission region, to understand the transport of PAHs to the Arctic. Dissolved PAHs showed significantly higher concentrations in the lower Great Lakes than those in the high Arctic. Concentrations of dissolved PAHs (Σ_{19}PAHs) ranged from 33 to 300 pg/L in the Fram Strait; the vertical profiles generally exhibited a decreasing trend toward deep waters, which was potentially influenced by hydrological and biogeochemical processes. PAHs were exported from the Arctic Ocean to the North Atlantic through the Fram Strait and the Davis Strait.

Plain Language Summary Polycyclic aromatic hydrocarbons (PAHs) are continuously released from multiple sources, mainly from combustion processes situated in populated and industrial regions. They are capable of long-distance transport to reach remote and deep oceans. To better understand PAH transport and fate, polyethylene passive samplers were deployed in deep waters of the Fram Strait, surface seawater of the Canadian Archipelago, as well as in air and surface water of the mid-latitude Great Lakes. Concentrations of dissolved PAHs in the lower Great Lakes were significantly higher than those in the high Arctic, indicating the presence of emission sources in the Great Lakes. The vertical profiles of dissolved PAHs generally exhibited a "surface enrichment and depth depletion" pattern in the Fram Strait, which was potentially affected by hydrological and biogeochemical processes. Pyrogenic sources were the dominant origin of PAHs in both the Arctic and Great Lakes, with some biomass burning sources possibly from wild fires in the sub-Arctic boreal forest regions. PAHs were exported from the Arctic Ocean to the North Atlantic, with small fluctuations of mass transport through the Fram Strait due to the continuous release of contaminants in mid-low latitude regions, as well as those primary and secondary sources within the Arctic.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of semivolatile organic compounds of great public concern because of their continuous release from pyrogenic and/or petrogenic sources, their ubiquitous distribution in populated and remote regions, as well as toxicity and potential carcinogenicity (Kim et al., 2013; Manzetti, 2013; Shen et al., 2013). The Arctic region receives PAHs through long range atmospheric and oceanic transport; the region is considered a "barometer" for global pollution (Balmer et al., 2019). Petrogenic sources of PAHs might also be important in the Arctic, given the contributions of coal/peat/bitumen deposit erosion by deglaciation and oil seeps (Lin et al., 2020; Yunker & Macdonald, 1995; Yunker et al., 2011). The expected decline of atmospheric PAHs in the Arctic due to the reduction of global emissions is offset by increased volatilization caused by global warming and increased wildfire (Yu et al., 2019). Understanding the relative importance of local and long range transport sources, as well as hydrological and biogeochemical processes of PAHs is essential, especially in a rapidly changing Arctic (Townhill et al., 2022).

Hydrologically, the highly stratified Arctic Ocean is a semi-enclosed deep basin surrounded by shallow continental shelves, with waterways connecting the Atlantic and Pacific oceans (Timmermans & Marshall, 2020). PAHs enter the Arctic Ocean with major and minor inflow of Atlantic and Pacific Water. Additionally, the occurrence of

PAHs in the central Arctic Ocean is strongly influenced by atmospheric inputs/outputs, freshwater discharge from large rivers and the transpolar ice drift (Ahmed et al., 2020; Elmquist et al., 2008; Ji et al., 2019; Ma et al., 2013). After being subjected to complex biogeochemical process, PAHs eventually leave the Arctic Ocean with cold and fresh waters via straits in the Canadian Arctic Archipelago and outflow through the Davis Strait, as well as in the East Greenland Current (EGC) on the west side of Fram Strait (Karpouzoglou et al., 2022; Zhang et al., 2016).

Biogeochemically, the photodegradation and biodegradation of the bioavailable PAHs, as well as the biological pump of PAHs associated with settling particles to deep waters, are viewed as the key processes in depleting PAHs in surface seawater, and further promote atmospheric PAHs deposition (González-Gaya et al., 2016, 2019; Liu, Chen, et al., 2021). While changes in the Arctic, including rising temperatures, declining sea ice coverage, and increasing riverine inputs may promote the release of PAHs from various deposits; these processes further impact their air-water exchange and transfer to deep waters (Ma et al., 2011; Pouch et al., 2017). Climate change has resulted in a "borealistion" of the Arctic, with increasing warming and influx of Atlantic Water (AW) (Wang et al., 2020). Meanwhile, some southern species, such as coccolithophore (*Emiliania huxleyi*), also expanding their northward boundaries, together with increasing primary productivity and changing food web structure, might also alter the hydrological, biogeochemical processes, and final fate of PAHs (Borgå et al., 2022; Fossheim et al., 2015; Lewis et al., 2020; Oziel et al., 2020).

Given the continuous release of PAHs and rapid climate change, although the occurrence and transport of PAHs were well documented in the Chukchi Sea and coastal Russia regions of the Arctic (Chen et al., 2021; Ji et al., 2019; Ke et al., 2017; Pouch et al., 2021), there is little work of PAHs in the Fram Strait, the only deep-water passage connecting the Arctic and Atlantic Ocean, or the Canadian Archipelago, the dominant surface-water passage from the Arctic to the North Atlantic. In this study, polyethylene (PE) passive samplers were deployed at deep mooring sites in the Fram Strait during 2014–2015 and 2018–2019, as well as in surface seawater of the high-latitude Canadian Archipelago and in both air and surface water of the mid-latitude lower Great Lakes during 2018–2019. Heavy urbanization and population along the shores of the lower Great Lakes make it a representative potential source for PAHs (McDonough et al., 2014). By combining results from these three locations, the objectives of this research were to (a) investigate the occurrence and potential sources of PAHs in high Arctic regions, as well as in comparison with the lower Great Lakes region. (b) provide knowledge of the vertical profiles of dissolved PAHs and possible influencing factors in their transport in remote deep ocean waters, and (c) estimate mass transport fluxes of PAHs through the Canadian Arctic Archipelago and the deep-water passage Fram Strait.

2. Materials and Methods

2.1. Sampling

According to the method described previously (Booij et al., 2002), 50 µm PEs were pre-spiked with performance reference compounds. The PEs were deployed at different water depths of deep moorings deployed in the eastern and western Fram Strait during 2014–2015 and 2018–2019 (31 samples), as well as in surface seawater of the Canadian Arctic Archipelago (6 samples), and in air and surface water (9 samples) of the lower Great Lakes during 2018–2019 (Figure 1, Figure S1 in Supporting Information S1). For atmospheric sampling, PEs were deployed at ~1–2 m height and fixed inside two inverted bowls to prevent rainfall and direct solar radiation. For water sampling, PEs were strung on stainless steel wires and attached to stainless steel cages. The surface-water cages were fixed to subsurface floats at ~4–5 m depth, while the deep-water cages were fastened to deep moorings at different depths for ~1 year. After recovery, the PE samplers were held at 4°C immediately, and kept frozen for shipment and storage until further pretreatment. The detailed sampling and hydrographic conditions are listed in Table S1, Text S1 and Figure S2 in Supporting Information S1.

2.2. Sample Analysis and QA/QC

The extraction, purification and instrument analysis method were conducted as described previously (McDonough et al., 2014). Briefly, all PEs were spiked with deuterated PAHs as surrogate standards and cold extracted in hexane twice overnight. The extracts were concentrated down to approximately 100 µL and spiked with p-terphenyl-d_{14} as an injection standard. Then, PAHs (Table S2 in Supporting Information S1) were analyzed using a 6890N GC (Agilent, USA)-Quattro Micro tandem mass spectrometer (Waters, Micromass, UK) with a DB5-MS fused silica
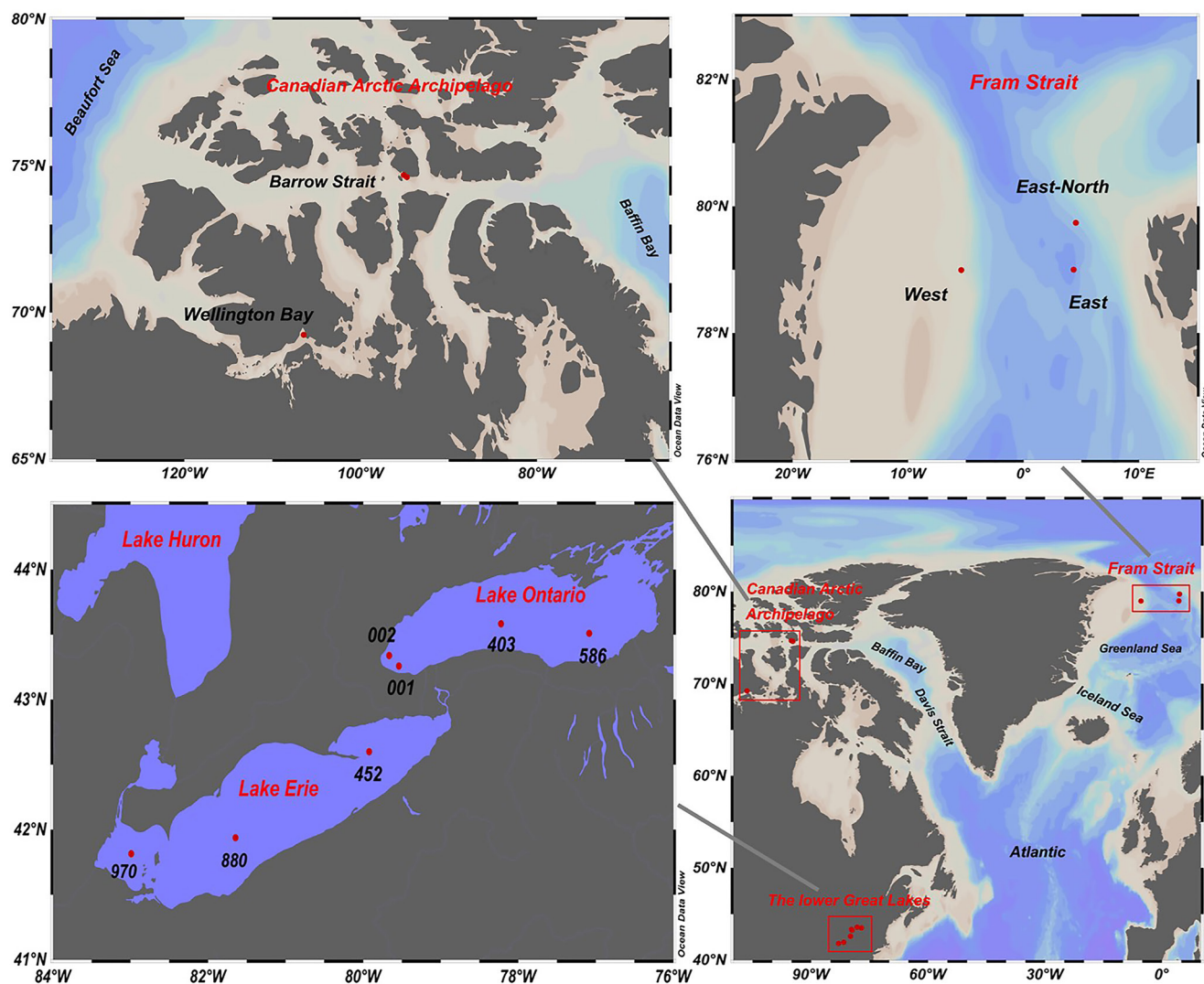
Figure 1. Sampling sites in the lower Great Lakes, Canadian Arctic Archipelago and Fram Strait during 2018–2019.

capillary column (30 m × 0.25 mm, Agilent J&W) in EI + multiple reaction monitoring mode. The methods for determination of sampling rates (Table S3 in Supporting Information S1), ambient air and freely dissolved water concentrations are described in detail in Text S2 in Supporting Information S1.

For each batch of samples, field and/or procedure blanks were included. Concentrations of all target compounds were recovery-corrected and blank-subtracted. The method detection limits were calculated as the mean plus 3 times the standard deviation (3σ) of field/procedure blanks (Table S4 in Supporting Information S1). The average recoveries of surrogate standards ranged from $58 \pm 7.5\%$ for d_{10}-phenanthrene to $99 \pm 18\%$ for d_{12}-chrysene (Table S5 in Supporting Information S1).

2.3. PAHs Mass Transport

To estimate PAHs mass transport through the Fram Strait, the mean PAH concentrations of different water masses were multiplied by the average water volume from boundary currents, which were defined based on prior work (Karpouzoglou et al., 2022; Stöven et al., 2016; Wang et al., 2020). The mean PAH concentrations of different water masses were derived based on the trapezoidal integration of the PAH concentrations through the water column. For PAHs mass transport estimates through the Davis Strait, the mean PAH concentrations were multiplied by the average water volume transport of currents (Zhang et al., 2016). The mean PAH concentrations in

the Nares Strait, Lancaster Sound, and Jones Sound were assumed the same as in the Barrow Strait. Detailed information for these calculations can be found in Text S2 in Supporting Information S1.

3. Results and Discussion

3.1. Depth Profiles of Dissolved PAHs in the Fram Strait

Detailed information on individual PAH concentration at different depths in the Fram Strait are listed in Tables S6 and S7 in Supporting Information S1. Total dissolved PAH concentrations (Σ_{19}PAHs) ranged from 33–300 pg/L and 60–210 pg/L during 2014–2015 and 2018–2019, respectively. PAHs generally displayed similar composition profiles for both deployment periods, dominated by the heavier four-ring, five-ring and six-ring PAHs, but few lighter three-ring PAHs (Figure 2, Figure S3 in Supporting Information S1). Specifically, four-ring PAHs accounted for 47%–94% of Σ_{19}PAHs, with fluoranthene (FluA) being the dominant compound. Moreover, the dissolved PAH concentrations during 2014–2015 were generally comparable with those detected during 2018–2019, especially in the western Fram Strait. This indicated constant outflow of PAHs from the Arctic through the EGC, probably reflecting the continuous deposition of atmospheric PAHs in the Arctic (Yu et al., 2019). The decrease of PAHs at the depth of ~400 m in the eastern Fram Strait from 2014–2015 to 2018–2019 by ~100 pg/L might indicate a slight reduction of PAHs inflow through West Spitsbergen Current.

The depth profiles of PAHs in the Fram Strait are shown in Figure 2. PAH concentrations generally decreased with increasing depth for both deployments, implying continuous external input of fresh PAHs, and degradation/loss in the water column. PAH levels at varied water depths were closely related to the water mass structure. Specifically, in the eastern Fram Strait, higher PAH concentrations were present in AW, likely due to the presence of primary PAH sources. During the northward transport process, the warm AW sinks when encountering with cold Polar Water (PW). Therefore, PAH concentrations were higher at ~400 m than those at ~200 m for both eastern sites of 2014–2015 and east-north sites of 2018–2019. In the western Fram Strait, relatively lower PAH concentrations were shown in Polar Surface Water at ~70 m depth, which might be due to the dilution effect of fresh water/sea ice melt water inputs in the Arctic Ocean; PAH concentrations increased at ~250 m depth as a result of mixing with Recirculating Atlantic Water (RAW). It is noticeable that PAHs generally displayed medium level in the Arctic Intermediate Water (AIW) at ~1,000 m depth, due to sinking of AW and photo-/bio-degradation during this process. Moreover, PAH concentrations were markedly lower in deep waters (>2,000 m) either for eastern or western Fram Strait, which was generally representative of old Eurasian Basin Deep Water and Greenland Sea Deep Water (EBDW/GSDW) with a mean age of 250 ± 30 years (Beszczynska-Möller et al., 2012; Stöven et al., 2016).

The compositional profiles of PAHs also varied with water depths (Figure 2, Figure S3 in Supporting Information S1). Specifically, three-ring PAH concentrations were extraordinarily low in old deep waters (>1,300 m), and demonstrated a depth-depletion pattern, probably due to microbial degradation. In contrast, contributions of four-ring, five-ring and six-ring PAHs generally increased owing to their strong binding to particles, settling to deep waters combined with the release of PAHs when particles were remineralized by bacteria at depth (as the PE samplers only sample freely dissolved PAHs) (González-Gaya et al., 2019).

The depth profiles of PAHs in this study were similar with those observed in the Mediterranean Sea (Dachs et al., 1997), the Black Sea (Maldonado et al., 1999), the Prydz Bay (Cai et al., 2016), the North Atlantic (Booij et al., 2014; Schulz-Bull et al., 1998; Sun et al., 2016), the Canary Basin (Booij et al., 2014) and the Chukchi Sea (Ke et al., 2017), all exhibiting a surface enrichment-depth depletion pattern (Table S8 in Supporting Information S1). Furthermore, some other contaminants observed in the Fram Strait, such as per- and polyfluoroalkyl substances (PFAS) also displayed a similar decreasing trend toward deep waters (Joerss et al., 2020), possibly due to their continued production and application. However, some legacy persistent organic pollutants, including polychlorinated biphenyls (PCBs) and organochlorine pesticides, showed different depth profiles, with higher concentrations in intermediate and/or deep waters (Ma et al., 2018), probably resulting from their worldwide production bans in the 1970s coupled with greater persistence in the water column compared to PAHs.
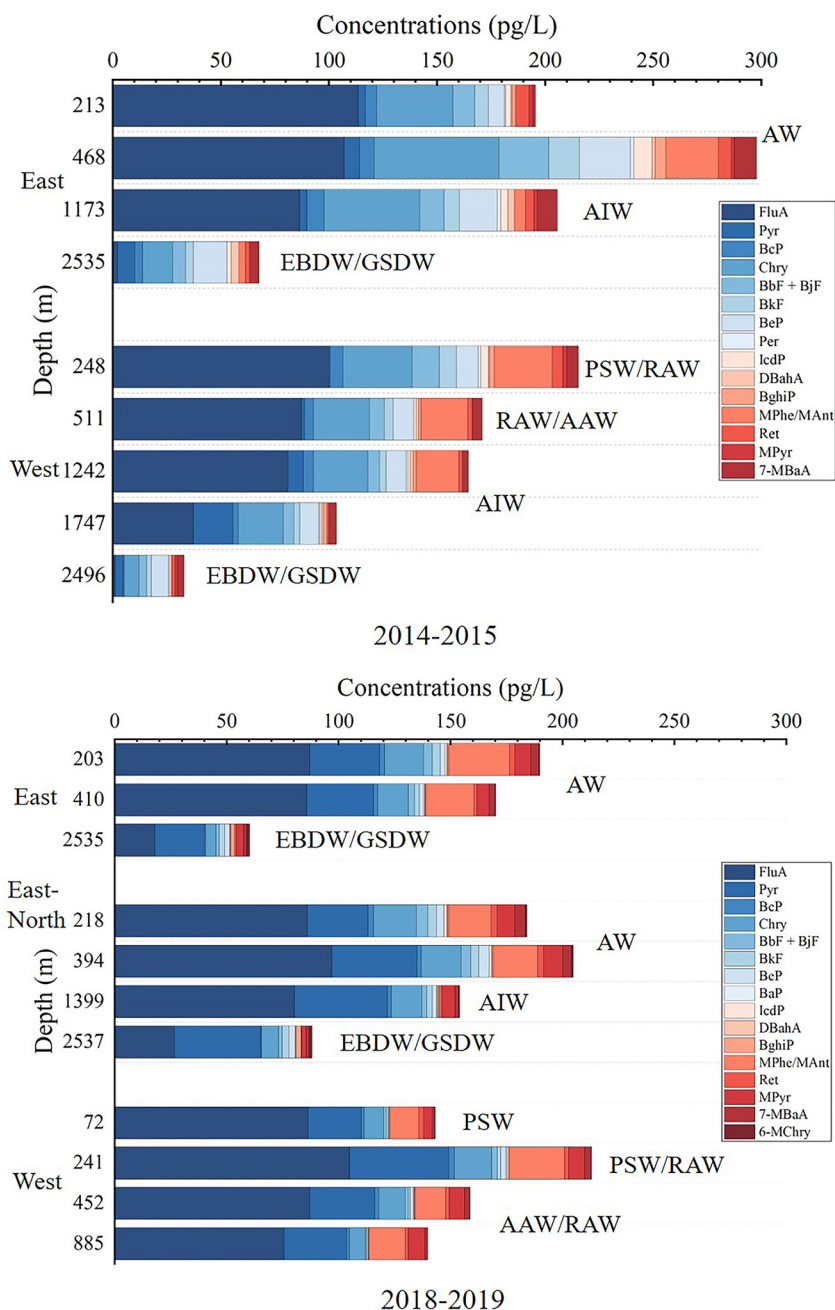
Figure 2. Depth profiles of dissolved Polycyclic aromatic hydrocarbons with a detection frequency >50% in the Fram Strait.

3.2. Occurrence of Dissolved PAHs in Surface Water of the Canadian Arctic Archipelago and the Lower Great Lakes

Detailed information on individual PAH concentration in surface water are listed in Table S9 in Supporting Information S1. For the Canadian Archipelago, total dissolved PAH concentrations in the surface water of coastal Barrow Strait and Wellington Bay were 1,100 and 640 pg/L, respectively. Their compositional profiles were dominated by three-ring PAHs at both sites (~70%) (Figure S4 in Supporting Information S1), consistent with previous studies of PAHs in surface seawater of remote oceans (Cai et al., 2016; González-Gaya et al., 2016). Concentrations of the dominant compounds acenaphthene (Ace, 310 and 210 pg/L), pyrene (Pyr, 240 and 170 pg/L) and phenanthrene (Phe, 160 and 70 pg/L) were also comparable with levels in the Arctic surface seawater reported previously (Chen et al., 2021). In contrast, the three-ring Ace and Phe were not detected in

the Fram Strait deep moorings, where PAHs had presumably undergone photo-/bio-degradation, preferentially removing lower weight compounds. Overall, compared with other studies (Table S10 in Supporting Information S1), dissolved PAH concentrations in surface water of the Canadian Arctic Archipelago were generally similar to those observed in the North Pacific in 2014 (1,800–16,000 pg/L, passive sampling) (Zheng et al., 2021) and in the Arctic Ocean in 2016 (820–3,700 pg/L, active sampling) (Chen et al., 2021), but lower than those detected in the Arctic fjords (200–310,000 pg/L, active sampling) (Pouch et al., 2021).

For surface water in the lower Great Lakes, concentrations of dissolved PAHs (Σ_{22}PAHs) ranged from 1,500 to 14,000 pg/L and averaged 6,200 ± 5,700 pg/L, which were significantly higher than those in the remote Canadian Archipelago ($p < 0.05$). The proximity to riverine discharges and wastewater treatment plants effluents were suggested to be responsible for such high levels of PAHs (McDonough et al., 2014), while the sites in the Canadian Archipelago were far from sources of air or water pollution. Similar to the Canadian Archipelago, the composition profiles were dominated by three-ring PAHs in the surface water of lower Great Lakes, with acenaphthylene (Acey) and Ace being the predominant compounds. This indicated that the Great Lakes region contains numerous emission sources that could transport PAHs to the Canadian Archipelago.

3.3. Gaseous PAHs and Air-Water Exchange Gradients in the Lower Great Lakes

The sum of gaseous PAHs (Σ_{21}PAHs) in the lower Great Lakes ranged from 1,000 to 3,900 pg/m^3, with a mean of 2,200 ± 1,000 pg/m^3 (Table S11 in Supporting Information S1). The gaseous PAHs showed especially high concentrations (Σ_{21}PAHs > 3,000 pg/m^3) at the sites close to populated regions of Lake Erie and Lake Ontario, which were greatly influenced by anthropogenic sources (McDonough et al., 2014). The predominant three-ring PAHs accounted for 87 ± 6.5% of PAHs composition profiles in the lower Great Lakes (Figure S5 in Supporting Information S1). The air-water fugacity ratios (f_a/f_w) of PAHs in the lower Great Lakes were >>1 (Table S12 in Supporting Information S1), indicating the process of atmospheric deposition and the lower Great Lakes acted as the regional sink of PAHs due to their continued emissions.

Although for this study, air passive samplers were not deployed in the Arctic region, the historical and recent data sets indicated great spatiotemporal variability of atmospheric PAHs in the Arctic. Specifically, the annual mean gaseous PAH concentrations (sum of 16 USEPA priority PAHs, Σ_{16}PAHs) were 170 and 500 pg/m^3 in the year of 2018 and 2019 at Zeppelin Station (Bohlin-Nizzetto et al., 2019, 2020), while significant higher levels of gaseous PAHs were present in the coastal Russia Arctic (4,300–4,600 pg/m^3) and from Chukchi Sea to the Canadian Basin (680–1,500 pg/m^3 in 2010, 670–3,900 pg/m^3 in 2014) (Ji et al., 2019; Ma et al., 2013; Zheng et al., 2021). Combined atmospheric level of PAHs in lower Great Lakes and high Arctic suggest not only the transport from the population/industrial centers further south, but also the existence of point sources of PAH within the Arctic and/or sub-Arctic regions.

3.4. Source Identification of PAHs

Molecular diagnostic ratios, principal component analysis followed by multiple linear regression analysis and positive matrix factorization (PMF) models have been widely applied to identify the source profiles and recognize quantitative contributions of different PAH sources in marine environments (Tobiszewski & Namieśnik, 2012; Zhang et al., 2021, X. Zhang, Zhang, et al., 2022b). Since parent- and alkyl-PAHs generally displayed significant and positive relationships ($p < 0.05$), indicating the same sources and similar environmental behavior (Figure S6 in Supporting Information S1), these compounds were analyzed together for source identification. The main results are presented below, and more detailed information is described in Text S2 and Text S3 in Supporting Information S1.

Two specific PAHs diagnostic ratios, FluA/(FluA + Pyr) and indeno (1,2,3-c,d)pyrene/(indeno (1,2,3-c,d)pyrene + benzo (ghi)perylene) (IcdP/(IcdP + BghiP)), were calculated to differentiate PAHs from pyrogenic (petrogenic combustion and grass/wood/coal combustion) and petrogenic origins. Specifically, the ratios of FluA/(FluA + Pyr) and IcdP/(IcdP + BghiP) ranged from 0.22 to 0.99 and 0.13 to 0.65 in the Arctic regions, and from 0.37 to 0.88 and 0.54 to 0.68 in the lower Great Lakes (Table S13 in Supporting Information S1). Besides rare petrogenic sources mainly in the Arctic samples, pyrogenic sources were the predominant PAHs sources for both Arctic and lower Great Lakes regions (Figure S7 in Supporting Information S1), in accordance with previous studies (Liu, Zheng, et al., 2021; Lohmann et al., 2009; Zheng et al., 2021). MDRs, however,

should be interpreted with caution due to different environmental processing of the isomers during atmospheric and oceanic transport, as well as photo-/bio-degradation (Galarneau, 2008).

For source apportionment, three significant factors were determined from PCA, which explained >85% of the total variability of the original dissolved PAH data set of the Canadian Archipelago, and results of the following MLR showed that contributions were 47% for fossil fuel and engine combustion sources, 28% for petroleum sources, and 25% for biomass/coal combustion sources, respectively. For seawater of the Fram Strait, three principal components accounted for >80% of the total variance, implying three sources: coal combustion, plus petroleum sources (50%), engine combustion (39%) and fossil fuel combustion (11%). For surface water of the lower Great Lakes, the extracted two principal components explained 100% of the total PAHs variance, while the estimated contributions were 62% for coal, fossil fuel and engine combustion sources, 38% for biomass combustion and petroleum sources (Table S15 in Supporting Information S1). Additionally, three significant factors were extracted by the PMF model to identify the source profile of atmospheric PAHs in the lower Great Lakes. Specifically, the potential sources were 34% from petroleum sources, 29% from biomass, coal and fossil fuel combustion sources, and 37% related to engine combustion sources (Figure S9 in Supporting Information S1). Overall, source identification results demonstrated gaseous and dissolved PAHs we detected from the lower Great Lakes and the Arctic were mainly derived from pyrogenic sources. The difference between the preponderance of biomass burning in the Arctic and fossil fuel emissions in the Great Lakes region can be explained by the resurgence of wild fires in the sub-Arctic boreal forest regions (Luo et al., 2020; Yu et al., 2019), but the presence of traffic, power generation and industrial activities relying on fossil fuels in the Great Lakes region.

3.5. Mass Transport of PAHs Through the Fram Strait and Davis Strait

For the Fram Strait, the resulting water mass transport of Σ_{17}PAHs during 2014–2015 were 31 ± 9.1 Mg per annum (Mg/a) in AW, 11 ± 0.84 Mg/a in PW and 19 ± 10 Mg/a in Recirculating and Arctic Atlantic Water (RAW/AAW), respectively, with a net northward transport fluxes of 1.5 ± 4.2 Mg/a from the North Atlantic to the Arctic Ocean (Figure 3b). For the year of 2018–2019, the estimated mass transport was 24 ± 5.3, 7.7 ± 2.3 and 17 ± 9.0 Mg/a in the boundary currents of AW, PW and RAW/AAW, respectively, with net southward transport fluxes of 0.74 ± 2.7 Mg/a from the Arctic Ocean to the North Atlantic (Figure 3d). These minor fluctuations indicated the close to steady state of mass transport of PAHs through the Fram Strait. The influxes of PAHs through AW were generally comparable with the annual atmospheric deposition fluxes of 16 parent PAHs during 1972–1992 to the Canadian Arctic and sub-Arctic regions (~37 Mg/a) (Peters et al., 1995). Compared with other compounds transported through the Fram Strait, the mass flows of some dominant PAHs of this study, such as FluA, chrysene (Chry) and Pyr, were in the same order of magnitude as α- and γ-hexachlorocyclohexanes (HCHs), perfluorooctanoic acid and chlorinated organophosphate esters (Cl-OPEs)-tris(1-chloro-2-propyl) phosphate (TCPP), but 1–2 orders of magnitude higher than PCBs and dichlorodiphenyltrichloroethanes (Joerss et al., 2020; Ma et al., 2018; McDonough et al., 2018).

It is noticeable that in both years, most five-ring and six-ring PAHs (benzo(b)fluoranthene (BbF), benzo(j)fluoranthene (BjF), benzo(k)fluoranthene (BkF), benzo(e)pyrene (BeP), perylene (Per), IcdP and BghiP) displayed net northward transport to the Arctic Ocean, however, the four-ring PAHs, including FluA and benzo(c)phenanthrene (BcP), generally exhibited net southward transport to the North Atlantic Ocean. The different contributions of atmospheric and oceanic transport of comparable lighter and heavier PAHs might explain the molecular weight-dependent net transport through the Fram Strait. The more volatile/lighter PAHs, which partly originate from emissions happening at lower latitudes, are prone to transport to the Arctic through faster long-range atmospheric transport, and deposition via air-water gas exchange, while the heavier PAHs, tend to be particle attached, might be more easily scavenged along the transport pathway and migrate to the remote Arctic through slower oceanic currents (Keyte et al., 2013). Moreover, the removal from the water column of the heavier PAHs via vertical particle settling, particularly in the shallower coastal shelf seas might also contributed to the reduced net southerly outflow from the Arctic of heavier PAHs (relative to the lighter PAHs). To a lesser extent, the longer retention of more hydrophobic/heavier PAHs in melting snow/sea ice might be another possible explanation for their reduced net southerly outflow, although large parts of the Arctic Ocean are now dominated by single season ice (rather than multi-year ice) (Kozhevnikov et al., 2021; Pawlak et al., 2021). The molecular weight-dependent net transport of PAHs we observed was similar with those for PFASs, as a net southward transport for short-chain PFAS, while a net northward transport for the PFASs with ≥8 perfluorinated carbon atoms (Joerss et al., 2020).
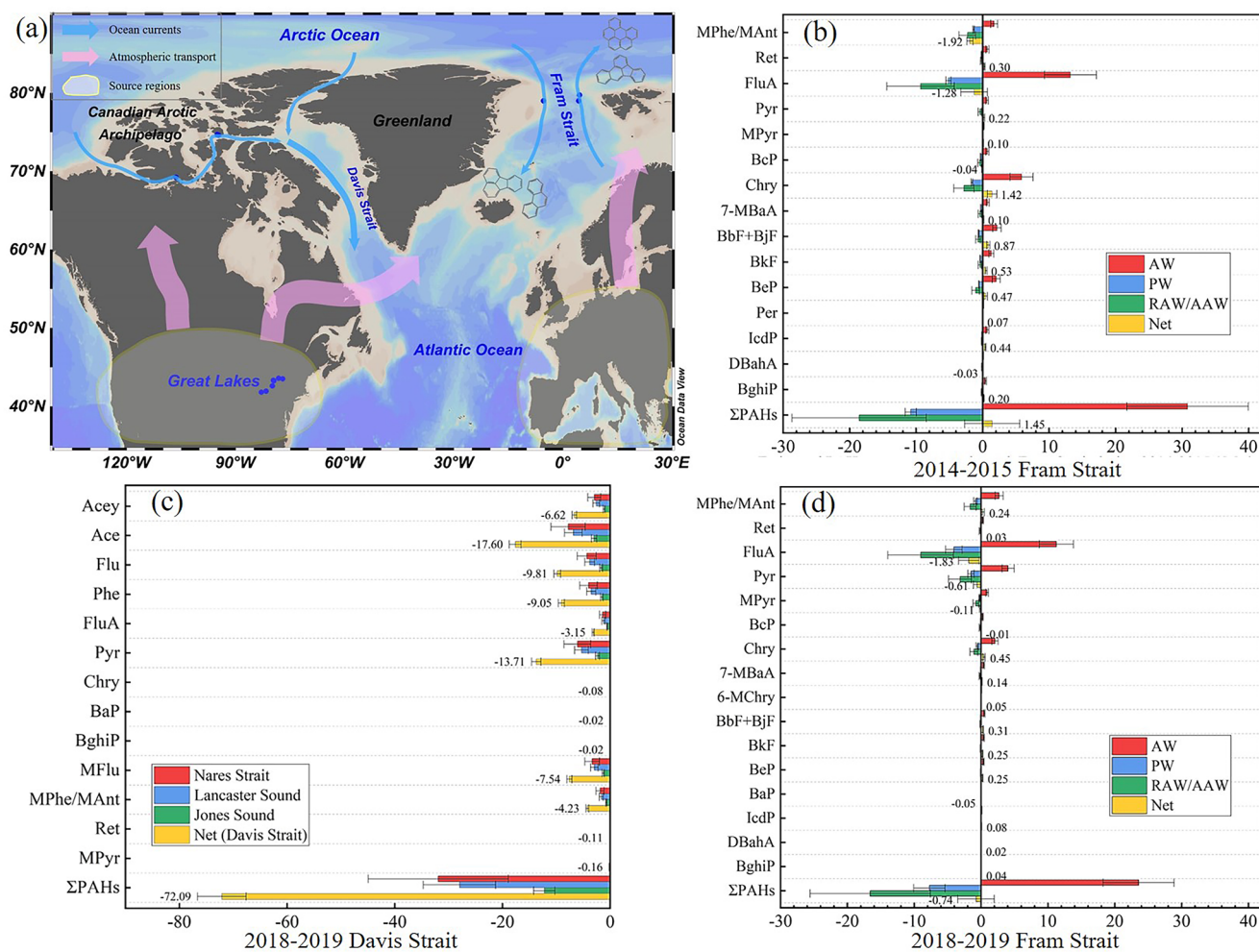
Figure 3. Conceptual feedback loop diagram of Polycyclic aromatic hydrocarbons (PAHs) in studied regions (a), and estimated PAHs mass transport (Mg/a) (PAHs with a detection frequency >50%) through the Fram Strait during 2014–2015 (b) and 2018–2019 (d) via boundary currents and through the Davis Strait during 2018–2019 (c). Positive values describe northward fluxes into the Arctic Ocean, whereas negative values describe southward fluxes to the North Atlantic Ocean.

Meanwhile, for the Davis Strait, the resulting water mass transport estimations of Σ_{14}PAHs during 2018–2019 were 32 ± 13 Mg/a in the Nares Strait, 28 ± 6.7 Mg/a in the Lancaster Sound and 12 ± 2.0 Mg/a in the Jones Sound, respectively, with a combined net southward transport fluxes of 72 ± 4.5 Mg/a through the Davis Strait from the Arctic Ocean to the North Atlantic Ocean (Figure 3c). Transport through the Davis Strait was 2–3 times as large as transport through the Fram Strait from the Arctic, although the ocean delivery of PAHs through the Fram Strait was almost balanced, by the counter flux of PAHs from the Arctic to the North Atlantic. We should also note that there would be inflows from the Atlantic to the Barents, which is not discussed in this study.

Our results suggested that although the Arctic Ocean is in the process of "borealistion" (Polyakov et al., 2020) and has continuously been impacted by PAHs influxes, to some extent, PAHs are released from the Arctic Ocean to the North Atlantic through ocean currents, showing the feedback effect of high latitude on mid-low latitude regions (Figure 3). The small fluctuations of mass transport of PAHs through the Fram Strait might be explained by the continuous release of contaminants in mid-low latitude regions, as well as those increase at high latitudes, including the frequent occurrence of northern boreal forest fires, as well as the secondary release due to melting snow/ice with warming tendency in the Arctic regions (Balmer et al., 2019; Luo et al., 2020; Wang et al., 2020; Yu et al., 2019; Zhao et al., 2019).

4. Conclusions

High levels of gaseous and dissolved PAHs in the lower Great Lakes, as well as air-water exchange gradients indicated this heavy urbanized and populated region represents both high emission sources for PAHs transport toward remote regions, but also as regional sinks of continuously released PAHs. The differences in concentrations of dissolved PAHs between the high-latitude Canadian Archipelago and the mid-latitude lower Great Lakes, together with the general "surface enrichment and depth depletion" pattern of dissolved PAHs in the Fram Strait, demonstrated the continuous inputs of fresh PAHs from populated to remote regions, as well as degradation during their long-range transport. The occurrence and transport of PAHs in the Arctic were jointly controlled by hydrological and biogeochemical processes. Specifically, PAH concentrations and composition profile at varied water depths were not only closely related to the water mass structure, but also potentially influenced by microbial degradation and the "biological pump." Source apportionment results indicated PAHs in both Arctic and Great Lakes were mainly derived from pyrogenic sources, with some representative of biomass burning sources possibly from wild fires in the sub-Arctic boreal forest regions.

The Arctic Ocean is in the process of "borealistion" and has continuously been impacted by PAHs influxes, while the mass transport of PAHs through the Fram Strait and the Davis Strait indicated the release of PAHs from the Arctic Ocean to the North Atlantic, showing the feedback effect of high latitude on mid-low latitude regions. The small fluctuations of mass transport of PAHs through the Fram Strait might be explained by the continuous release of contaminants in mid-low latitude regions, as well as those primary and secondary sources within the Arctic.

Data Availability Statement

PAH concentration data of deep water in the Fram Strait during 2014–2015 and 2018–2019, and of surface seawater in the Canadian Arctic Archipelago and of both air and surface water in the lower Great Lakes during 2018–2019 are available online in Zenodo (at https://doi.org/10.5281/zenodo.7352264) (Zhang, Ma, et al., 2022). The data and figures are also available in the Supporting Information S1.

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