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Review

Legacy and emerging organic contaminants in the polar regions

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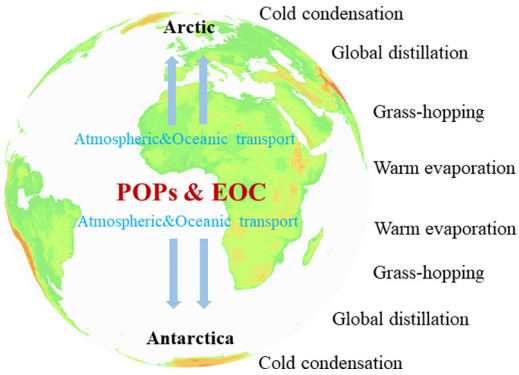
HIGHLIGHTS

- Legacy POPs have shown declining temporal trends in the Arctic and the Antarctica.
- EOCs are widely present in various environmental matrices in polar regions.
- Increasing human activities in polar regions caused the local discharge of EOCs.
- Climate change induces remobilization of POPs and EOCs in polar regions.
- Many EOCs have become of an emerging concern for the polar ecosystems.

GRAPHICAL ABSTRACT



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ABSTRACT

The presence of numerous emerging organic contaminants (EOCs) and remobilization of legacy persistent organic pollutants (POPs) in polar regions have become significant concerns of the scientific communities, public groups and stakeholders. This work reviews the occurrences of EOCs and POPs and their long-range environmental transport (LRET) processes via atmosphere and ocean currents from continental sources to polar regions. Concentrations of classic POPs have been systematically monitored in air at several Arctic stations and showed seasonal variations and declining trends. These chemicals were also the major POPs reported in the Antarctica, while their concentrations were lower than those in the Arctic, illustrating the combination of remoteness and lack of potential local sources for the Antarctica. EOCs were investigated in air, water, snow, ice and organisms in the Arctic. Data in the Antarctica are rare. Reemission of legacy POPs and EOCs accumulated in glaciers, sea ice and snow may alter the concentrations and amplify their effects in polar regions. Thus, future research will need to understand the various biogeochemical and geophysical processes under climate change and anthropogenic pressures.

Abbreviations: AMAP, Arctic Monitoring and Assessment Programme; BFR, Brominated flame retardant; dl-PCBs, dioxin-like PCBs; EOCs, Emerging organic contaminants; ImPACT, Input Pathways of persistent organic pollutants to AntarCTic; LCCPs, Long-chain chlorinated paraffins; LRAT, Long-range atmospheric transport; LRET, Long-range environmental transport; MCCPs, Medium-chain chlorinated paraffins; OCPs, Organochlorine Pesticides; OPEs, Organophosphate esters; PAHs, Polycyclic aromatic hydrocarbons; PBDEs, Polybrominated diphenyl ethers; PCBs, Polychlorinated biphenyls; PCNs, Polychlorinated naphthalenes; PFASs, Per- and poly fluoroalkyl substances; POPs, Persistent organic pollutants; PPCPs, Pharmaceuticals and personal care products; SC, Stockholm Convention; SCAR, Scientific Committee on Antarctic Research; SCCPs, Short-chain chlorinated paraffins.

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Contents

1. Introduction

Pristine polar ecosystems have beein impacted by numerous natural and anthropogenic factors. The combination of increasing human activities and changing climate causes major pressure for the polar environment (AMAP, 2017; AMAP, 2021). Direct and indirect inputs of man-made pollutants from local emissions and remote continental sources led both the Arctic and Antarctica to act as ultimate sinks, which can cause unexpected disturbances and pose risks to polar ecosystems, creating a global problem for the Earth in the future.

Along with the rapid increase in global pollution and the rapid development of the economy and industry, a variety of chemical pollutants have been introduced into the environment in large quantities (Brown and Wania, 2008; Cabrerizo et al., 2013; Policy, 2017; Wania, 2007). Persistent organic pollutants (POPs) are synthetic chemicals and have common characteristics of persistence, toxicity and bioaccumulation potential and the tendency of long-range environmental transport (LRET) (Arnot et al., 2011; Nash et al., 2017). Therefore, POPs are considered to pose a threat to the environment and human health and are subject to the Stockholm Convention to reduce and ultimately eliminate these compounds from the environment (SC, 2022a). Initially, twelve POPs were recognized as causing adverse effects on humans and the ecosystem (Table S1). Subsequently, sixteen new chemicals were added to the Stockholm Convention in the following years (SC, 2022a). The seventeenth meeting of the POPs Review Committee (POPRC-17) to the Stockholm Convention has adopted decisions on six chemicals or groups. Among them, chlorpyrifos, chlorinated paraffins with carbon chain lengths in the range C14–17 and chlorination levels at or exceeding 45% by weight, and long-chain perfluorocarboxylic acids (PFCAs), their salts and related compounds were moved to next stage of review. UV-328 and Dechlorane Plus were requested fur review, and methoxychlor should be listed under Annex A to the Convention (Table S1) (SC, 2022b).

In addition to classic POPs, a number of organic chemicals have been found in polar regions as emerging organic contaminants (EOCs). Generally, emerging contaminants are expected to be chemicals that show potential to pose risks to human health or the environment but are not yet subjected to international regulatory (Sauve and Desrosiers, 2014). EOCs include alternative brominated flame retardants (BFRs), short-chain poly- and perfluoroalkyl substances (PFASs) and their precursors, organophosphate esters (OPEs), phthalate esters (PAEs), chlorinated paraffins (CPs), pharmaceuticals and personal care products (PPCPs), UV filters (UV-Fs) and cyclic volatile methyl siloxanes (cVMSs), and their transformation products (AMAP, 2017).

Organic contaminants exist both in water, vapor phase of air or attached to particles. They can be transported from source regions to remote areas via atmosphere and oceanic circulations (Nash, 2011). These chemicals can finally reach the polar regions through global distillation and fractionation processes (Wania, 2007; Wania and Dugani, 2003). Therefore, both the Arctic and Antarctica have accumulated large quantities of organic contaminants and become important sinks for global POPs. Furthermore, human activities in polar regions have changed and are rapidly increasing, such as intensive expeditions, blooming of polar tourism and shipping. For instance, cruise passenger numbers for the high Arctic have increased from around 50,000 in 2005 to 80,000 in 2016 (Taylor et al., 2020). In the Antarctic, the visitor number reached about 73,991 in summer of 2019/2020 (IAATO, 2020). Direct release of organic contaminants from the research stations was observed in both the Arctic and Antarctica (Kwok et al., 2013; Wild et al., 2015). Thus, assessing and monitoring local source contributions of recently used novel organic chemicals in polar regions will be of increasing importance (Nash, 2011; Wild et al., 2015). As terrestrial animals, marine mammals and circumpolar Inuit in the Arctic are highly affected by organic contaminants because of geographic locations, lipid-rich diet, and at the top of food chains (Nash, 2011).

The Arctic Environmental Protection Strategy, signed by eight Arctic countries (Canada, Denmark, Finland, Iceland, Norway, Russia, Sweden the United States) in 1991, proposed to monitor the levels of anthropogenic pollutants and assess their effects in all Arctic environmental compartments as organized by the Arctic Monitoring and Assessment Programme (AMAP). To date, AMAP has published several important assessment reports for POPs and EOCs in the Arctic (AMAP, 2002; AMAP, 2017; AMAP, 2021). Chemical monitoring has become a basic requirement of the Protocol on Environmental Protection to the Antarctic Treaty (the Madrid Protocol) adopted in 1991, which was established to limit the long-term impacts of direct human activities on the Antarctic environment (www.ats.aq/e/protocol.html). The working group of Input Pathways of Persistent Organic Pollutants to Antarctica (ImPACT) under the Scientific Committee on Antarctic Research (SCAR) was proposed to facilitate coordinated investigation and monitoring of chemical input to the Antarctica region. The research activities and findings for POPs in the Antarctica have been reported (Nash, 2011; Vecchiato et al., 2015a; Kallenborn et al.,

2013; Pozo et al., 2017; Bhardwaj et al., 2018). The organic contamination found in the Antarctic mainly reflects the emissions of the Southern Hemisphere, which has a relatively low population and historical industry (Nash, 2011; Kallenborn et al., 2013).

In this report, we review the levels of both classic POPs and EOCs in biotic and abiotic environmental matrices in polar regions, which include PFASs, PBDEs, OPEs, organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), CPs, polychlorinated naphthalenes (PCNs) and selected PPCPs. In addition, studies on atmospheric deposition, air-water exchange and remobilization of EOCs were summarized to better understand the environmental pathways of EOCs in polar regions. Consequently, research gaps and future perspectives are identified for important research and monitoring efforts required for assessing the impact of organic contaminants in the changing polar regions.

2. Environmental levels and profiles of POPs and EOCs in polar regions

2.1. Polybrominated diphenyl ethers (PBDEs)

From the group of BFRs existing data are mostly from legacy PBDEs, already listed as POPs (Table S1). Atmospheric PBDE levels from station- and

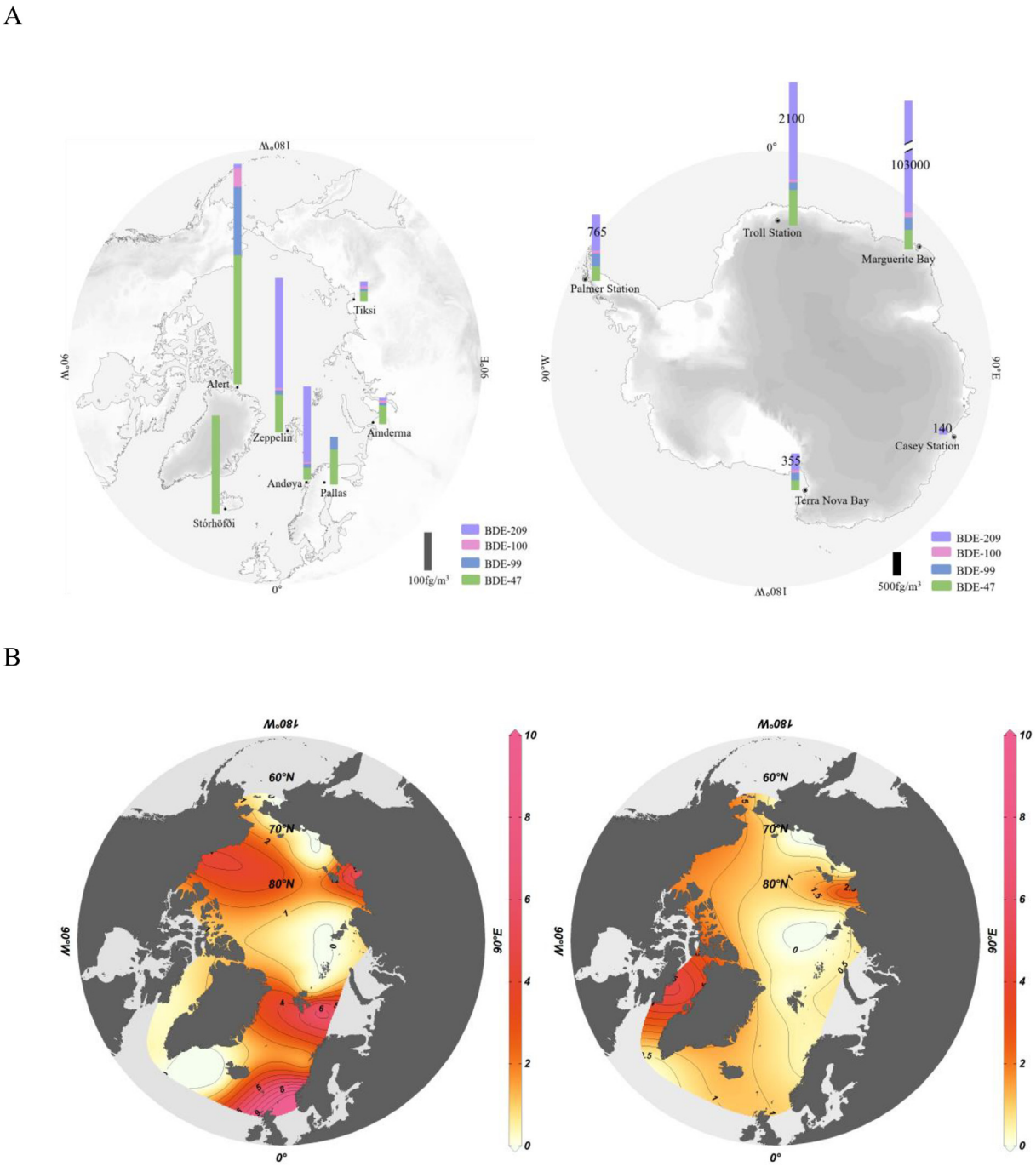

Fig. 1. (A). Concentrations of PBDEs measured at 6 research stations in the Arctic (left) and at 4 stations in Antarctica (right). BDE-209 is the predominate BDEs at all Antarctic stations, followed by BDE-47, BDE-99 and BDE-100 (Dickhut et al., 2012; Nash et al., 2021; Salvado et al., 2016). In the Arctic, BDE-209 is the major BDE in air at Zeppelin and Andøya, while BDE-47 dominants at other 4 stations; (B). PBDEs (pg/L) in surface seawater from the Arctic (left: the sum of BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190; Right: BDE-209) (Salvado et al., 2016).

ship-based measurements in polar regions are summarized in Table S2. Air monitoring under the AMAP shows that PBDE concentrations at most stations were declining (Hung et al., 2016), responding to effective regulation and hence a reduction in technical penta- and octa-BDE release from North America and Europe in the mid-2000s. Some PBDE congeners (e.g., BDE-47, 99, and 100) exhibited a strong cyclical pattern, with a maximum observed in summer, when revolatilization of these congeners likely controlled the atmospheric levels (Fig. 1A). However, the time trend of air BDE-209 levels in the Arctic is still considered disputable (Wong et al., 2021).

Through an air survey of PBDEs in West Antarctica from 2011 to 2017, no clear temporal trend was observed, with BDE-28 and BDE-47 dominating the PBDE air profile and a high detection rate of BDE-183 (Hao et al., 2019). BDE-209 was the predominant BDE, followed by BDE-47, BDE-99 and BDE-100 (Dickhut et al., 2012; Nash et al., 2021) (Fig. 1A). A higher level was presented at Casey Station than at the other sites, which might be influenced by local emissions (Khairy et al., 2016; Nash et al., 2021). The observations highlighted the need for longitudinal monitoring for the determination of robust temporal trends in the Antarctica.

PBDEs in water and snow from marine and terrestrial environments of polar regions are summarized in Table S3. Snowmelt concentrations of PBDEs in continental and coastal surface snow ranged from 130 pg/L to 340 pg/L, with BDE-47 and BDE-99 contributing the most (Vecchiato et al., 2015a). These concentration ranges and composition profiles agreed well with other observations in the Antarctica (Khairy et al., 2016). BDE-209 showed predominance in snow pits from the Devon Ice Cap (680–100,000 pg/L) and in ice cores from Holtedahlfonna (Meyer et al., 2012; Hermanson et al., 2010). Concentrations of PBDEs were about 1 pg/L in the Russian rivers Ob and Yenisei (Carroll et al., 2008) and ranged from n.d. to 21.4 pg/L in Canadian Arctic lakes (McDonough et al., 2018). PBDEs in the polar mixed layer throughout the Arctic Ocean ranged from 0.3 to 11.2 pg/L, with higher concentrations found in the pan-Arctic shelf seas (Salvado et al., 2016; Moeller et al., 2011a; Moeller et al., 2011b) (Fig. 1B). PBDEs in surface sediments have been measured in Arctic/Antarctic marine and terrestrial environments with concentrations from not detected to hundreds ng/g (Cai et al., 2012a; Corsolini et al., 2019; Jiao et al., 2009; Ma et al., 2015). In addition to long-range atmospheric transport (LRAT) driven PBDE enrichments in soils of polar regions, PBDE accumulations related to release from research stations are also of concern (Vecchiato et al., 2015b).

PBDE enrichments in biota are provided in Table S4. Studies on PBDE in biotic samples, including phytoplankton, krill, fish, and fur seal milk, from the Antarctica over 14 years (2000–2014) illustrated a dominant contribution made by BDE-47 and 99 as well as an increasing tendency of PBDE concentrations in fur seal milk, krill, and phytoplankton (Markham et al., 2018). This contrasts with a general decreasing trend in the Northern Hemisphere. Time-series investigations of marine mammals, seabirds, marine and freshwater fish and blue mussels in the context of AMAP showed a typical trend of increasing concentrations up to the mid-2000s for BDE-47, followed by a decrease in concentration (Rigét et al., 2019). However, penta-, octa-, and deca-BDE concentrations in Canadian Arctic belugas between 1997 and 2013 showed no significant trend, suggesting that the effect of global PBDE regulations has yet to be perceived (Simond et al., 2017).

2.2. Polycyclic aromatic hydrocarbons (PAHs)

Atmospheric PAHs in polar regions have been investigated either by long-term site-based observations or ship-based expeditions during various sampling periods (Table S5); stations of Alert, Zeppelin, and Pallas have been covered by the AMAP for Arctic PAH monitoring since 1992, 1994, and 1996, respectively (Kong et al., 2014). The concentrations of PAHs showed clearly declining trends from the North Pacific to the Arctic (Fig. 2A), suggesting LRAT from the Asian continent to the Arctic (Zheng et al., 2021). Based on a survey at the station Alert in Canada from 1993 to 2002, the concentrations of 16 PAHs which are on the United States Environmental Protection Agency (US EPA) priority pollutant list declined from 1992 to 1996, but the concentrations of gaseous PAHs increased in

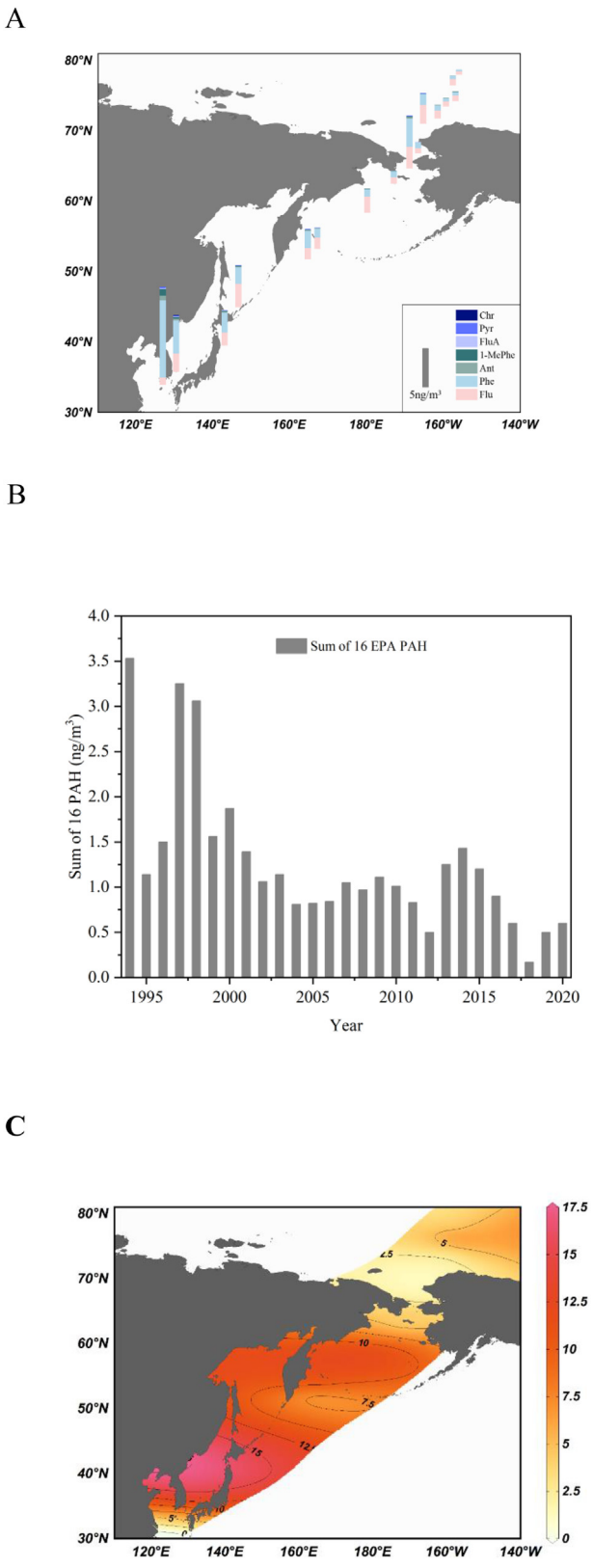

Fig. 2. (A). Spatial distribution of PAHs from the North Pacific to the Arctic (Zheng et al., 2021); (B). Annual mean concentrations of the sum of 16 EPA PAHs measured at Zeppelin station, Arctic from 1994 to 2020 (Bohlin-Nizzetto et al., 2014, 2015, 2017, 2018, 2019, 2020, 2021; Bohlin-Nizzetto and Aas, 2016); (C). Distribution of PAHs in seawater from the North Pacific and the Arctic (Zheng et al., 2021).

1998 (Balmer et al., 2019; Yu et al., 2019). However, the annual mean concentrations of the sum of 16 EPA PAHs measured at Zeppelin showed deceasing trend from 1998 to 2020 (Fig. 2B). Similar declining trend of PAHs was observed in Antarctic air from 2013 to 2019 (Na et al., 2020a). The varying trends of PAH concentrations present in the air at different stations reflect geographic dynamics of the natural and anthropological sources. Ice retreat, ship emission and forest fire in the high latitudes can cause enhanced PAHs levels in polar regions (Lammel et al., 2015; Luo et al., 2020; Vecchiato et al., 2015a). Besides, pollution with PAH derivatives in the Arctic boundary layer displayed six fold and two orders of magnitude higher levels of oxy-PAHs than parent and nitro-PAHs, respectively, with their highest levels observed in the spring (Drotikova et al., 2021).

Snow and ice cores are important preservative sinks for PAHs in polar regions (Table S6). PAHs in surface snowmelt from the high Arctic substantially varied spatially from several to thousands ng/L, reflecting main contributions from their LRAT (Lebedev et al., 2018; Masclet et al., 2000; Vecchiato et al., 2018). PAH levels of the same order of magnitude were encountered in Antarctic snow (Lebedev et al., 2018; Vecchiato et al., 2015a; Xie et al., 2020). Through the retrieval of time-dependent PAH pollution profiles in ice cores from polar regions, some historical epochs were responsible for depth variability in PAH levels (Giannarelli et al., 2017; Masclet et al., 2000; Vehviläinen et al., 2002).

PAH levels in surface seawater varied in magnitude from picogram to nanogram per liter, depending on sampling period and location (Table S7). Fig. 2C shows distribution pattern of PAHs in the North Pacific and the Arctic. Depth profiles of seawater PAH concentrations revealed a general decrease in PAH abundance with depth, indicating PAH depletion in the deep sea (Giannarelli et al., 2017). Studies on PAHs in the Arctic rivers mainly focused on the Mackenzie River, with total PAHs up to several hundred ng/L (Table S7) (Yunker et al., 2002). PAHs ranged from n.d. to >1000 ng/L in remote lakes of the Arctic (Lehmann-Konera et al., 2020), and from 10 to 50 ng/L in the lake water of the Grovnes and Broknes Peninsula, East Antarctica (Bhardwaj and Jindal, 2020; Bhardwaj et al., 2021).

Sediment PAHs in the bottom boundary layer or cores from the Arctic/Antarctic Ocean are compiled in Table S8, with relatively high PAH loadings found in the coastal area. Based on investigations in sediments of the Barents Sea in the 1990s and 2000s, PAH levels showed no significant changes with time. The concentrations of PAHs in sediment cores ranged from 2.0 to 27 ng/g from Deception Island (DCP) and from 13 to 60 ng/g in Penguin Island (PGI), in the South Shetland Archipelago, Antarctica (Sutilli et al., 2019), which are about 10 times lower in comparison to those measured in sediment cores from the Southwestern Atlantic continental shelf (45 to 305 ng/g) (Timoszczuk et al., 2021). Petrogenic-derived PAHs were predominant in the upper layer of the sediment core at Deception Island, suggesting influence of tourism and research stations activities in Antarctica. PAH levels in terrestrial soils at the Arctic/Antarctic sites are given in Table S8. As soil PAHs mainly occur through atmospheric scavenging-driven particle deposition, PAHs are up to dozens of μg/g in polar regions (Marquès et al., 2017). Generally, soils in the Antarctica, which are relatively more isolated from direct human influence, were less PAH contaminated than Arctic soils.

PAH bioaccumulation in organisms has received extensive attention in polar regions in recent decades (Table S9). OH-PAHs (PAH metabolites) are considered excellent chemical proxies for exposure to the parent PAHs in fish. In polar areas, PAH occurrences in multiple species have been reported, such as fish, invertebrates, penguins, seabirds (including eggs), and mussels, excluding marine mammals to our knowledge (Almeda et al., 2021; Montone et al., 2016; Szczybelski et al., 2016; Yu et al., 1995), from which it is noted that invertebrates at low trophic positions accumulate a larger amount of parent PAHs than their predators at higher trophic positions.

2.3. Organochlorine pesticides (OCPs)

The atmospheric OCPs concentrations generally declined on the basis of a 25-year long research at AMAP air monitoring stations, in which the target compounds included α- and γ-hexachlorocyclohexane (HCH), α-endosulfan, chlordane, dichlorodiphenyltrichloroethane (DDT) and its metabolites dichlorodiphenyldichloroethylene (DDE), hexachlorobenzene (HCB), drins (dieldrin, aldrin and endrin), endosulfan and heptachlor. The temporal trends of HCB, HCH isomers, DDTs and chlordanes from 1993 to 2020 at Zeppelin were illustrated in Fig. 3A, and the variations of all 6 Arctic stations were summarized by Wong et al. (2021). The reduction on global emission of technical HCHs and Lindane caused that α- and γ-HCH concentrations declined significantly in Arctic atmosphere. Similar to HCHs, DDT and DDE generally declined with some short-term stable periods. The ratios of DDE to DDT demonstrated that DDTs in the Arctic air mainly associated with the residual of aged technical DDTs. The seasonal variation of some OCPs (including HCHs, Chlordane and DDTs) with winter minimum and summer maximum was reported at some stations, indicating temperature played a critical role in some OCPs compounds at some stations. However, the seasonality of γ-HCH showed the controversy trend in other stations, suggesting the direct atmospheric transport was likely a source of HCHs therein (Wong et al., 2021). Generally, the OCPs in the Arctic air have the comparable half-lives, ranging from several to decades years (Wong et al., 2021).

The OCPs concentration followed an order of HCB > endosulfan-I > α-HCH > endrin > dieldrin in the eastern Antarctica atmosphere, according to a multiyear monitoring effort from 2010 to 2015 (Fig. 3B). HCB was the dominant OCPs compound measured both in the Arctic and Antarctic atmosphere. The average α-HCH concentrations were over 2 orders of magnitude higher in the Arctic than in the Antarctica. During the five years, no significant temporal tendencies of HCB and α-HCH were discerned. Moreover, the seasonality of austral summer maximum and austral winter minimum was also not consistent with those in Arctic atmosphere, suggesting the concentrations of HCB and α-HCH did not show temperature dependence in east Antarctica (Nash et al., 2017).

Mean concentrations of all OCPs in water-based matrices (i.e., seawater, sea ice, and snow) in the Arctic were generally higher than in the Antarctica (Bigot et al., 2017). HCH isomers (α-, β-, and γ-HCH) were detected in seawater, sea ice, and snow in the Arctic, whereas only γ-HCH was found in all these matrices in Antarctica, with α-HCH being found in seawater only. Generally, α-HCH was the predominant OCP in seawater and sea ice, followed by dieldrin in polar regions (Bigot et al., 2017). It was reported that *p,p'*-DDT was the dominant OCPs in the lakes of Broknes peninsula at Larsemann Hills area, East Antarctica, taking an order: ΣDDTs > endosulfan > drins > HCHs. The OCPs concentrations in lake waters were relatively higher than those in seawater (Bhardwaj et al., 2019; Bhardwaj and Jindal, 2020).

Among the selected OCPs measured in the Arctic char, toxaphene always showed the highest concentrations (1.6 to 170 ng/g wet weight (ww)), followed by ΣDDTs (0.54 to 61 ng/g ww) and ΣHCHs (0.03 to 4.24 ng/g ww). Most legacy OCPs showed a declining trend, which can be attributed to international restrictions on uses and emissions in circumpolar and neighboring countries. The OCP contaminant levels were among the highest reported in Arctic seabird species (Braune, 2007; Miljeteig et al., 2009). Chlordane, DDT, dieldrin and mirex showed significant decline or no significant change between 1975 and 2003 in the seabirds from the Canadian Arctic (Braune, 2007). However, significant increases were observed for ΣHCH in kittiwakes and fulmars as well that β-HCH and cis-chlordane in murres and fulmars. OCP concentrations varied in polar bears from different regions (Lie et al., 2003). OCPs have been widely detected in organisms collected from Antarctica, including Limpet, Antarctic cod, Amphipods, Antarctic icefish, Gentoo and Chinstrap penguins, Kelp gull, and South polar skua (Kim et al., 2021).

2.4. Polychlorinated biphenyls (PCBs)

The PCB concentrations in the air of the Arctic were in the range of < LOD to a few ng/m^3, which was much higher than those in the Antarctic air except for the Great Wall station (Hung et al., 2016; Wang et al., 2017; Wong et al., 2021). Among the POP monitoring stations in the Arctic, the

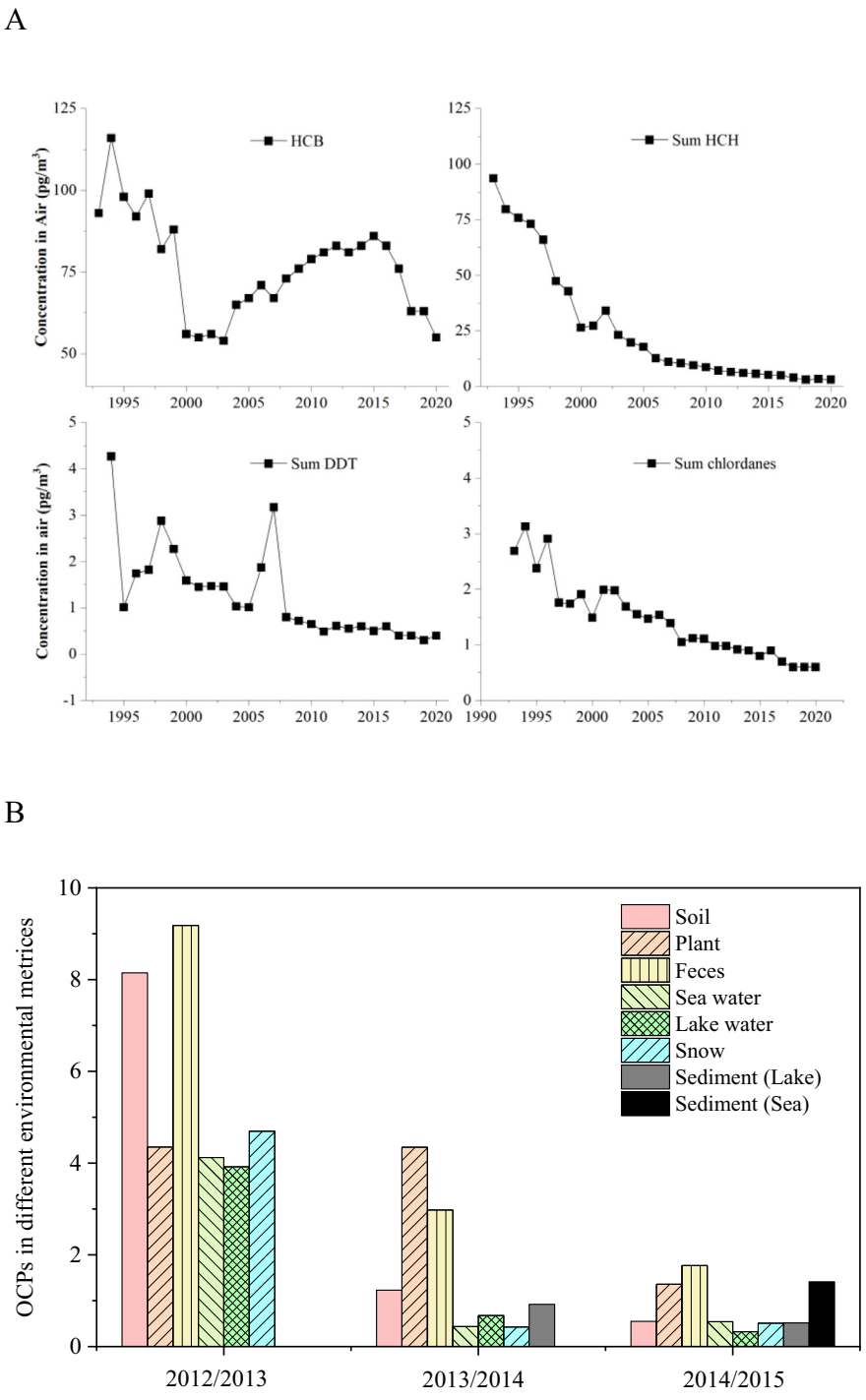

Fig. 3. (A). Temporal trends of annual atmospheric concentrations HCB, sum HCH, sum DDT and sum chlordane measured at Zeppelin station from 1993 to 2020 (Bohlin-Nizzetto et al., 2014, 2015, 2017, 2018, 2019, 2020, 2021; Bohlin-Nizzetto and Aas, 2016). (B). Distribution of OCPs in various environmental matrices in George Kind Island, Antarctica from 2012 to 2015.

highest level was determined in Zeppelin (Fig. 4a). Air concentrations of PCB-28, 52, 101, 118, 138, 153, 180 exhibited declining trends at most AMAP stations from 1992 to 2018 (Wong et al., 2021). It was predicted that PCB-180 would be one of the first two depletions from the Nordic atmosphere before 2020, and then the remaining PCBs would be depleted between 2020 and 2025 (Anttila et al., 2016). However, the apparent half-lives of PCBs, ranging from 6.1 to 35 years, demonstrated that PCBs remained in Arctic atmosphere for decades years (Wong et al., 2021). The PCBs derived from European continents posed influence on the seasonal variation of PCBs at different stations. Accordingly, the winter minimum and summer maximum were not observed at all stations and in all the PCB congeners' variation patterns as well (Wong et al., 2021). Fig. 4e shows the annual mean concentrations of 32 most toxic PCB congeners (Σ32PCBs) and 7 PCBs used as indicators in Europe (Σ7PCBs) from 2001 to 2020 (Bohlin-Nizzetto et al., 2014, 2015, 2017, 2018, 2019, 2020, 2021; Bohlin-Nizzetto and Aas, 2016). Overall, a decreasing trend presented from 2001 to 2020, while increase trend was observed from 2011 to 2016. The annual mean concentrations of PCBs were quite comparable

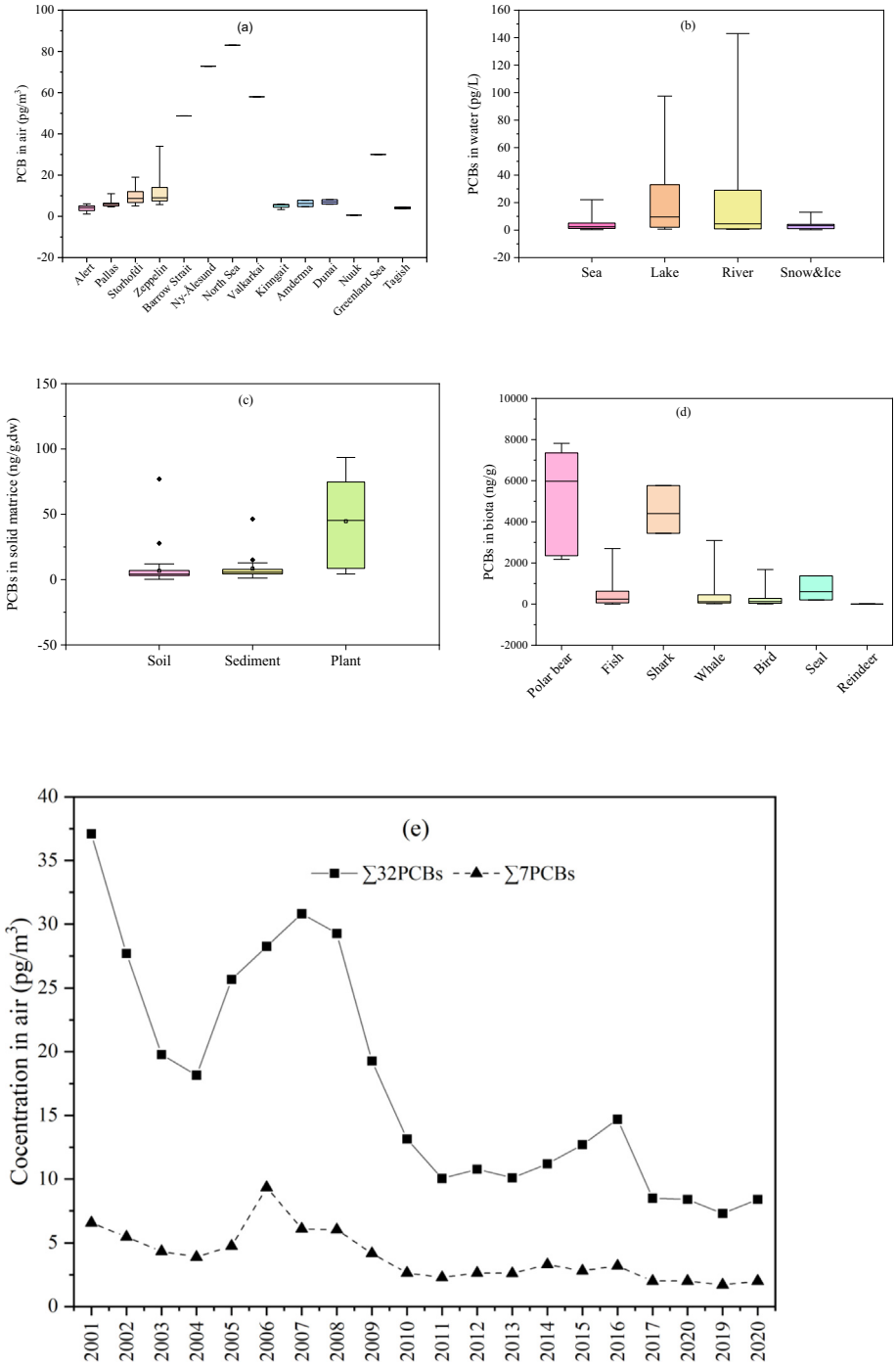

Fig. 4. (a-d). The PCBs concentrations in air, water, solid matrices and organisms in the polar region; (e). Annual mean concentrations of Σ32PCBs and Σ7PCBs (pg/m^3) in air samples at Zeppelin station from 2001 to 2020 (Bohlin-Nizzetto et al., 2014, 2015, 2017, 2018, 2019, 2020, 2021; Bohlin-Nizzetto and Aas, 2016).

in the years 2017–2020. A comparison between available data indicates that concentrations of PCBs in the Antarctic atmosphere have decreased since the 1980s (Bargagli, 2008).

PCBs in the aquatic environment of the Arctic areas, including seawater and lake water, were quite similar, ranging from < LOD to a few hundred pg/L (Sobek and Gustafsson, 2004; Pouch et al., 2021) (Fig. 4b). Similarly, PCB concentrations in the Arctic were higher than those in the Antarctica (Mangano et al., 2017). However, there were remarkable differences in PCB concentrations in snow and ice in the polar regions. Generally, the PCB concentrations in snow and ice were 1–2 orders of magnitude higher than those in seawater and lake water. PCB-11 was determined in the surface snow collected from Lomonosovfonna, Svalbard, accounting for 4% of the total PCBs (Garmash et al., 2013).

PCBs in surface sediment and sediment cores were investigated in marine areas of polar regions (Hong et al., 2012; Zhang et al., 2013; Combi et al., 2017; Deng et al., 2020a; Deng et al., 2020b). Concentrations of PCBs in Antarctic sediments were usually low but still higher than those in samples from low-impact and remote environments (Combi et al., 2017), suggesting the existence of local sources (Fig. 4c). PCBs in soil were reported in the range of 2.8–11 ng/g from Ny-alesund (Zhang et al., 2014) and 8.0–34 ng/g from King George Island, West Antarctica (Park et al., 2010). Enriched PCBs in vegetation such as lichens and mosses

(range 0.1–35 ng/g dw) were generally several orders of magnitude higher than those in polar abiotic matrices (Zhang et al., 2014).

PCB concentrations showed different trends in Arctic biota (Fig. 4d). A significant decrease was observed in seal blubber Holman between 1972 and 2016 (Houde et al., 2019), and PCBs decreased 4–11% per year in the lives of arctic foxes (*Vulpes lagopus*) (Andersen et al., 2015). The PCB concentrations in the blubber of male Antarctic minke whales (20–25 years old), *Balaenoptera bonaerensis*, were much lower than those in common minke whales, *Balaenoptera acutorostrata*, from the Northern Hemisphere (Yasunaga et al., 2015).

2.5. Chlorinated paraffins (CPs)

CPs are widely used as flame retardants and plasticizers in industrial and household products and include very-short-chain CPs (C < 10, vSCCPs), short-chain CPs (C10-C13, SCCPs), medium-chain CPs (C14-C17, MCCPs) and long-chain CPs (C > 18, LCCPs) according to the length of the carbon chain (Meziere et al., 2020). SCCPs officially entered the new POP list and were detected in the Arctic and Antarctica. CPs have been included in measurement campaigns at the Arctic atmospheric monitoring stations of Alert, Zeppelin, Barrow, Storhoefði and Little Fox Lake. The annual mean concentrations of SCCPs and MCCPs measured at Zeppelin showed that SCCPs were relatively constant, while MCCPs increased from 23 to 750 pg/m^3 from 2013 to 2020 (Fig. 5A). SCCPs were detected in air samples on King George Island, Antarctica, with concentrations ranging from 71.4 and 4230 pg/m^3 from 2014 to 2018 (Fig. 5B) (Jiang et al., 2021). An increasing temporal trend was observed for CPs in the Antarctica.

CPs in the Arctic biota were recently reviewed by Vorkamp et al. (2019a). Briefly, CPs have been investigated in freshwater fish from the Arctic with concentrations ranging from 0.12 to 1000 ng/g in lake trout, Arctic char, white fish, and Ninespine stickleback (Li et al., 2017a; Vorkamp et al., 2019a; Yuan et al., 2019). CPs up to C29 were detected in marine organisms from the Arctic, indicating long-range transport of LCCPs (Yuan et al., 2021). In Antarctica, SCCPs have been investigated in biota samples collected from the Fildes Peninsula at King George Island and Ardley Island, with concentrations ranging from 3.5 to 260 ng/g (Li et al., 2016). Casa et al. (2019) determined SCCPs in Southern Hemisphere humpback whales feeding in the Antarctic waters with concentrations up to 46 ng/g. In terms of carbon chain length, short-chain (C10) congeners comprised the main components in the Antarctic and Arctic samples, accounting for 56% and 49% of the total SCCPs, respectively (Li et al., 2017a; Li et al., 2016), which is consistent with the pollution characteristics of SCCPs in the Arctic marine mammals (Tomy et al., 2000).

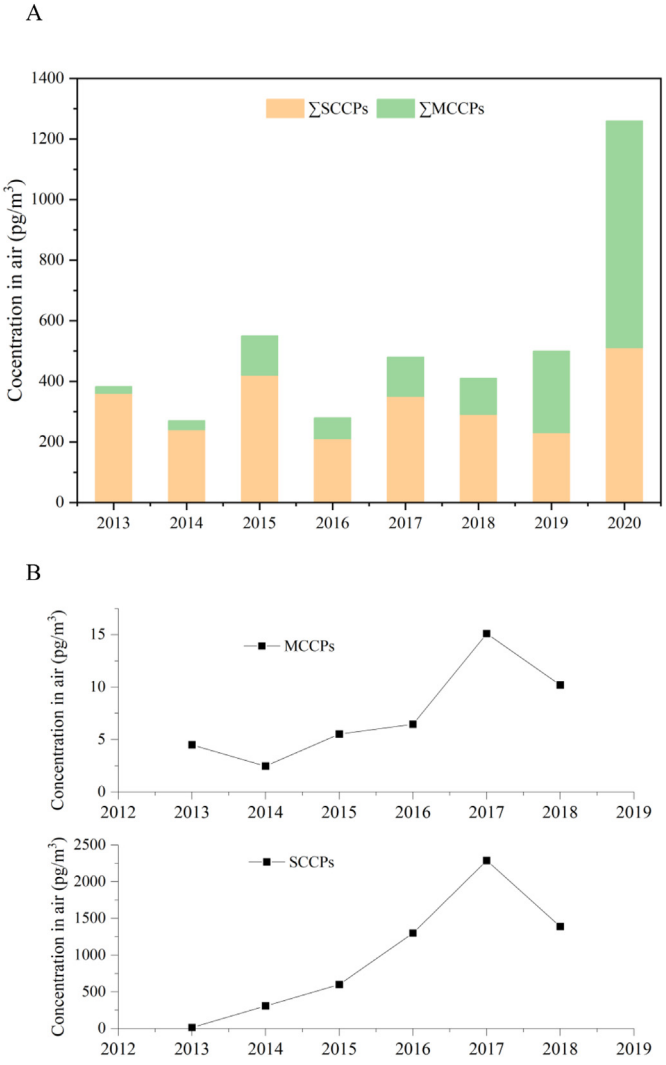

Fig. 5. (A). Concentrations of SSCCP and SMCCP (pg/m^3) (annual means and standard deviations) in air samples (gaseous and particle phase) at Zeppelin station, 2013–2017 (Bohlin-Nizzetto et al., 2014, 2015, 2017, 2018, 2019, 2020, 2021; Bohlin-Nizzetto and Aas, 2016); (B). Concentrations of SCCPs and MCCPs (pg/m^3) (annual means) in air samples (gaseous and particle phase) at the Great Wall Station (Georgia King Island, Fildes Peninsula of Antarctica), 2013–2018 (Jiang et al., 2021; Ma et al., 2014).

2.6. Polychlorinated naphthalenes (PCNs)

Studies of PCNs in polar regions have been reviewed by Bidleman et al. (2010). PCNs were detected in the air of the Arctic, with high concentrations (27–48 pg/m^3) present in Ny-Ålesund (Herbert et al., 2005), 8.7–47 pg/m^3 in Tromso, and 40 pg/m^3 in the Barents Sea (Harner et al., 1998). The annual average concentrations of PCNs of Alert, Dunai and Tagish are 0.69, 0.82 and 0.38 pg/m^3, respectively (Fig. 6). PCNs showed seasonal trends in Alert and Dunai. PCN concentration was higher in winter during the haze stage when the air mass originated mainly from the Eurasia (Helm et al., 2004). In Antarctica, the concentrations of PCNs were measured in atmospheric particles collected from Terra Nova Bay, ranging from n.d.-0.6 pg/m^3 (Fig. 6) (Barbaro et al., 2016). In addition, PCNs were also detected in the snow from Ny-alesund (350 pg/L) and Tromso (240 pg/L) in the Arctic (Herbert et al., 2005), indicating wide occurrence of PCNs in the Polar regions.

PCNs were determined in thick billed crow eggs collected from Prince Leopold Island in the Canadian Arctic between 1975 and 2014 with concentrations decreasing significantly at an average annual rate of 15 pg/g wet weight (ww) (Braune and Muir, 2017). High concentrations of PCNs were detected in marine mammals in the Arctic, such as polar bears (12–57 ng/g ww), seals (16–71 pg/g lw), and whales (36–380 pg/g lw) (Helm et al., 2002; Letcher et al., 2018; Wang et al., 2007). The concentration of PCNs was positively correlated to the age, sex, and blubber thickness of the male ringed seals (Wang et al., 2007). In the Ross Sea in Antarctic, PCNs were found in krill biomass at a concentration of 1.5 pg/g ww (Corsolini et al., 2002). The concentrations of PCNs in penguin samples have increased from 0.69 to 2.1 ng/g lipid weight (lw) to 0.012–26 ng/g lw (Kim et al., 2021). Relatively constant concentrations of PCNs were present in scallop tissue samples from 1996 to 2009 (Grotti et al., 2016).

2.7. Organophosphorus esters (OPEs)

OPEs have been widely detected in the atmosphere of the Arctic, Antarctica and adjunct oceans (Fig. 7A) (Xie et al., 2022). The median concentration of Σ_{13}OPEs was 50 pg/m^3 in the Canadian Arctic, 334 pg/m^3 in Longyearbyen and 74–852 pg/m^3 in Ny-alesund (Bohlin-Nizzetto et al., 2021; Han et al., 2020). Furthermore, OPEs have been measured in both gas and particle phases along transects with concentrations from 35 to 343 pg/m^3 in the European Arctic (Li et al., 2017b) and 232 to 1884 pg/m^3

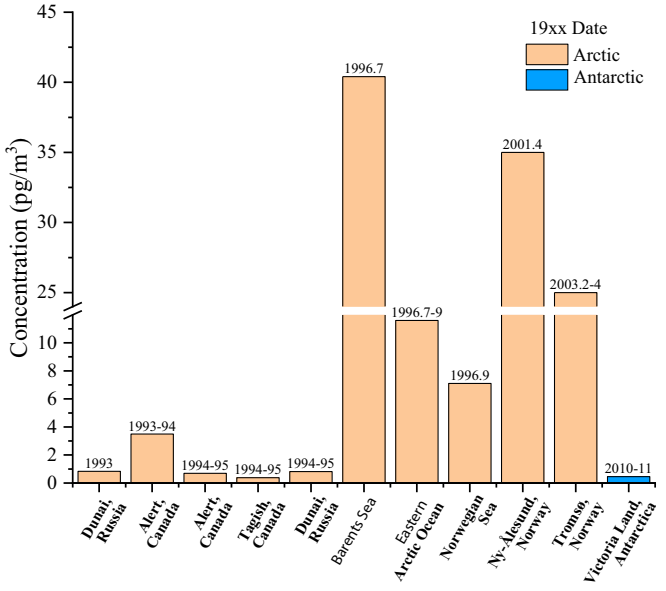

Fig. 6. Mean concentrations of PCN (pg/m^3) in air samples (gaseous and particle phase) in the Arctic and Antarctica.

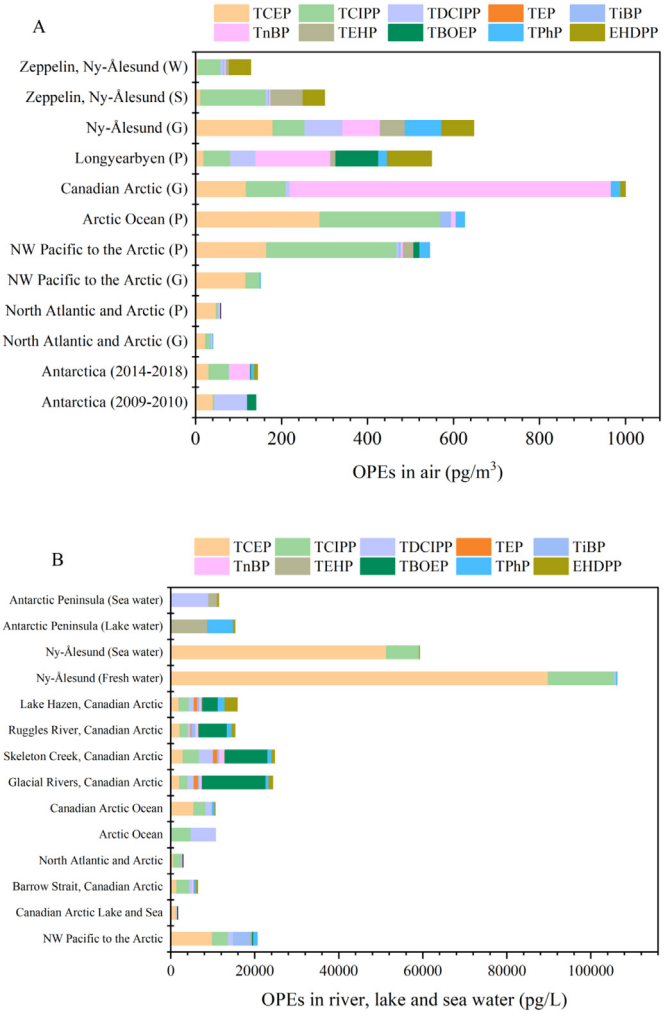

Fig. 7. Concentrations of OPEs (pg/m^3) (means) in air (A) and river, lake and glacier's water and seawater (B) in the Arctic and Antarctica.

in the northwestern Pacific to the Arctic Ocean (Na et al., 2020b). (Fig. 7A). In the Antarctica, the concentrations of OPEs ranged from 6.0 to 141 pg/m^3 in ship-bound particle samples (Cheng et al., 2013b) and from 33.9 to 404 pg/m^3 in the Western Antarctic Peninsula (Wang et al., 2020). Among the OPEs, chlorinated OPEs such as tris (2-chloroethyl) phosphate (TCEP) and tris (1-chloro-2-propyl) phosphate (TCIPP) are the predominant compounds, and Tributyl phosphate (TnBP) and tri-isobutyl phosphate (TiBP) are the major nonchlorinated OPEs.

OPEs have been investigated in seawater of the global ocean (Fig. 7B) (Xie et al., 2022). The concentrations of OPEs ranged from 8.5 to 143 ng/L (Σ_{11}OPEs) in the North Pacific to the Arctic (Na et al., 2020b), 0.35 to 8.4 ng/L (Σ_8OPEs) in the North Atlantic and East Greenland Sea (Li et al., 2017b), and 1.3 ($\Sigma_{non\text{-}Cl}$-OPEs) to 10 ng/L (Σ_{Cl}-OPEs) in the Canadian Arctic (Suhring et al., 2021). Comparable levels were reported with passive water samples (Gao et al., 2019; McDonough et al., 2018). Relatively high OPE concentrations were reported in the fjord of Ny-alesund (range: 8.7–358 ng/L, mean: 89 ng/L) (Gao et al., 2020), the Fildes Peninsula (<5.0–44.4 ng/L) (Gao et al., 2018), and in freshwater from the northern Antarctic Peninsula (19.6–9209 ng/L) (Esteban et al., 2016), suggesting input sources around the research stations (Choi et al., 2020). The concentrations of OPEs in the snow were 7.2 to 20.5 ng/L from Dome Concordia (Xie et al., 2020) and 0.05 to 2.0 ng/L for TCEP on the Antarctic ice sheet (Cheng et al., 2013a), which is in line with the levels detected in the Arctic snow (4.4–11 ng/L) (Li et al., 2017b) and in Canadian Arctic Lake Hazen and its tributaries (15–25 ng/L) (Sun et al., 2020). OPEs have been detected in oceanic sediment with concentrations of ΣOPEs ranging from 0.16 to 4.7 ng/g dry weight (dw) from the North Pacific to the Arctic (Ma et al., 2017), 0.12 to 57 ng/g dw in the Canadian Arctic (Suhring et al., 2021), and 0.01 to 15 ng/g dw on the coast of Ny-alesund (Gao et al., 2020). TCEP and TCIPP are the most abundant detected OPEs in ocean sediments.

In the Arctic biota, 10 OPEs were found in capelin, whereas only individual OPEs were present in other species, such as the kittiwake, brünnich's guillemot, glaucous gull, ringed seal, harbor seal, arctic fox and polar bear (Hallanger et al., 2015). Relatively high concentrations of ΣOPEs were observed in Kittiwakes (mean ΣOPEs: 666 ng/g lw) from Svalbard (Evenset et al., 2009) and in the liver of glaucous gulls from the eastern Canadian Arctic (mean ΣOPEs: 488 and 847 ng/g lw), respectively (Verreault et al., 2018). Five out of 17 OPEs were quantifiable at ng/g levels in polar bear fat samples (Letcher et al., 2018). Fu et al. (2020) investigated OPEs in biota from the Antarctic Peninsula and found OPE concentrations of a few ng/g.

2.8. Per- and polyfluorinated alkyl substances (PFASs)

PFASs have been widely determined in environmental matrices, including seawater, surface water, snow, ice, sediment and air, from polar regions, suggesting the long-range transport potential and persistence of PFASs in polar regions (Kwok et al., 2013; Nash et al., 2010; Xie et al., 2015; Yamashita et al., 2008; Young et al., 2007). Besides, the effluent of wastewater from research stations and local settlements can act as important local contamination source for PFASs in coastal areas (Wild et al., 2015). Neutral PFASs determined in the environmental samples include 6:2, 8:2, and 10:2 fluorotelomer alcohol (FTOH); 6:2, 8:2, and 10:2 fluorotelomer acrylates (FTAs); methyl and ethyl perfluorooctane sulfonamides (MeFOSA and EtFOSA); and methyl and ethyl perfluorooctane sulfonamidoethanols (MeFOSE and EtFOSE) (Cai et al., 2012b; Dreyer et al., 2009; Wang et al., 2015).

The neutral PFASs in the Southern Ocean and along the Antarctic Peninsula ranged from 3.7 to 48 pg/m^3 (Del Vento et al., 2012; Wang et al., 2015). 8:2 FTOH (mean: 12.7 ± 9.8 pg/m^3) was the most predominant compound (Fig. 8A). In the Arctic, neutral PFASs have been determined in the air samples collected during expedition cruises and from research stations, e.g., Alert, Villum, and Ny-alesund (Muir et al., 2019; Shoeib et al., 2006). The average Σ_7PFAS determined in the atmosphere at Villum ranged from 1.82 to 32.1 pg/m^3 (Bossi et al., 2016). Similar levels

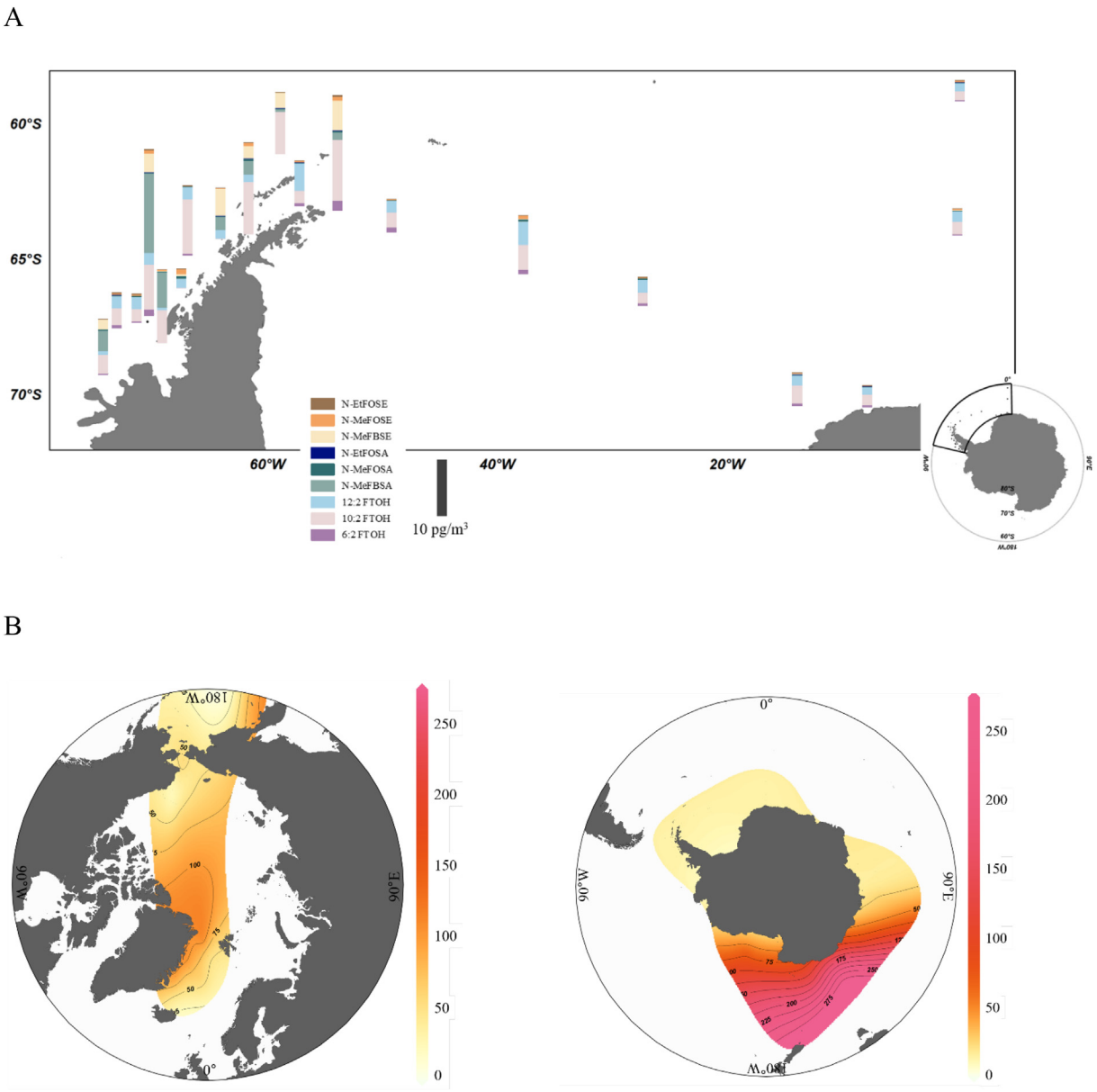

Fig. 8. (A) Neutral PFASs present in air from the Southern Ocean and Antarctic Peninsula (Del Vento et al., 2012; Wang et al., 2015); (B) Spatial distribution of PFOA (pg/L) in seawater of the Arctic (Left) and the Southern Ocean (Right) (Cai et al., 2012a, 2012b, 2012c, 2012d; Shan et al., 2021a, 2021b; Yamazaki et al., 2021; Zhao et al., 2012).

were reported in the atmosphere of Ny-Alesund (Xie et al., 2015) and in the Canadian Arctic (Wong et al., 2018). FTOHs showed clear seasonal trends, while the seasonal trends of FOSEs, FOSAs and FTAs were not remarkable because of their low levels.

Ionic PFASs are likely partitioned to the particle phase in the atmosphere owing to their physicochemical behaviors. At Alert Station, perfluorobutanoic acid (PFBA) (median: 0.097 pg/m^3) was the most detected compound in the air, followed by PFOS (median: 1.7 pg/m^3) and PFOA (median: 0.07 pg/m^3) (Wong et al., 2018). The annual mean PFOA in 2020 is 0.12 pg/m^3 at Birkenes, which is ~2 times higher than that of Andøya (0.05 pg/m^3) and Zeppelin (0.07 pg/m^3) (Bohlin-Nizzetto et al., 2021). Other ionic PFAS, such as perfluorodecanoic acid (PFDA), perfluorononanoic acid (PFNA), perfluoroheptanoic acid (PFHpA), perfluorohexanoic acid (PFHxA), perfluorobutane sulfonate (PFBS), perfluorodecane sulfonate (PFDS) and 6:2 fluorotelomersulfonate (6:2 FTS), were close to the method detection limits (MDLs). In the Southern Ocean, 12 PFAS were detected in air ranging from 1 to 5 pg/m^3 (Yamazaki et al., 2021), and short chain PFCAs such as PFBA, perfluoropentanoic acid (PFPeA), and PFHxA were the predominant PFASs.

Ionic PFASs have been determined in surface snow and ice cores from Longyearbyen and Ny-Alesund in Svalbard, Greenland and the Canadian Arctic (Kwok et al., 2013; Maclnnis et al., 2019; MacInnis et al., 2017). PFBA, PFOA, and PFNA dominated in ice-core samples, and PFOA was the main PFAS in surface snow. In Antarctica, PFASs have been measured in surface snow samples from Livingston Island, King George Island and inland on the Antarctic Plateau (Xie et al., 2020; Wang et al., 2015; Casal et al., 2017; Cai et al., 2012c). Hexafluoropropylene oxide dimer acid (HFPO-DA) was found in the Antarctic snow ranging from 4.7 to 13 pg/L (Xie et al., 2020). The mean concentration of Σ_{15}PFASs was 193 pg/L in ice melting lakes of Larsemann Hills, East Antarctica (Shan et al., 2021b). 6:2 chlorinated polyfluorinated ether sulfonate (6:2 Cl-PFESA) was reported in the lake water of Antarctica and ranged from <11–15 pg/L.

PFAS has been determined in the lake water from the Canadian Arctic, Faroe Island and Svalbard (Eriksson et al., 2013; Kwok et al., 2013; Stock et al., 2007). Lescord et al. (2015) determined perfluoro-4-ethylcyclohexanesulfonate (PFECHS) and fluorotelomer sulfonates (4:2-, 6:2-, and 8:2 FTSs) in Resolute and Meretta Lakes. Skaar et al. (2019) measured PFASs in the lake and run-off water from firefighting

training stations (FFTSs) in Longyearben and Ny-alesund. Ali et al. (2021a) found PFOS (19.09 ± 0.87 ng/L) and 6:2 FTS (1.46 ± 0.08 ng/L) in the FFTS-creek.

There are a few surveys for PFASs in oceanic seawater from the Southern Ocean and the Antarctic coast, with concentrations ranging from tens to hundreds pg/L (Casal et al., 2017; Yamazaki et al., 2021; Ahrens et al., 2010; Shan et al., 2021a; Zhao et al., 2012). In the Arctic Ocean, PFASs were measured in the North Pacific to the Arctic Ocean with a mean of 500 ± 170 pg/L (Cai et al., 2012d), which is higher than those in the Greenland Sea (ΣPFAS: 48 to 270 pg/L) (Zhao et al., 2012). PFOA is usually the predominant PFAS in the Arctic and the Southern Ocean with concentrations ranging from 10s to 100 s pg/L (Fig. 8B). Joerss et al. (2020) detected HFPO-DA in seawater of the Norwegian Sea ranging from <6.0 to 70 pg/L. PFBS (23–64 pg/L) was the predominant PFSA, while PFOS was below the MDL in most seawater samples. Garnett et al. (2021) found that PFOA was the dominant PFCA in near-surface seawater, with a mean concentration of 928 ± 617 pg/L in the Barents Sea (81°N) (Garnett et al., 2021), which is even higher than those recently measured in surface waters of the North Sea receiving the industrial discharge (Joerss et al., 2020). The vertical profiles showed higher PFAS concentrations in the surface water layer than in intermediate waters and negligible intrusion into deep waters (Joerss et al., 2020; Yeung et al., 2017).

PFASs have been measured in oceanic sediment in the Bering Sea and Chukchi Sea (Lin et al., 2020b). No clear temporal trends were observed in the sediment cores from 1975 to 2015. Emerging PFASs such as PFECHS, 3H-perfluoro-3-[(3-methoxypropoxy)propanoic acid] (ADONA), 8-chloro-perfluoro-1-octane sulfonic acid (8-Cl-PFOS), 6:2 and 8:2 chlorinated polyfluorinated ether sulfonate (6:2 Cl-PFESA and 8:2 Cl-PFESA) were detected in the Arctic lakes in Canada (MacInnis et al., 2019). 6:2 FTS (4.0 ng/g) was found in sediment samples in Svalbard (Ali et al., 2021b). Relatively high concentrations of PFASs (up to 1140 ng/g dw) were found in soil near FFTS (Skaar et al., 2019). Presently, few data are available for both neutral and ionic PFASs in sediment from Antarctica and the Southern Ocean.

Generally, relatively low concentrations of PFAS were present in the Antarctic food web (Giesy and Kannan, 2001; Tao et al., 2006; Schiavone et al., 2009). PFOS and perfluoroundecanoic acid (PFUnDA) were shown to be the predominant PFAS in blood samples of most species in the Antarctic region (Midthaug et al., 2021; Nash et al., 2010; Routti et al., 2015). Higher concentrations of PFAS were often found in the blood plasma or liver of migratory birds (Munoz et al., 2017). However, the report of elevated levels of PFAS in south polar skua during the breeding season implied that bioaccumulation of PFAS can occur through the food chain (Garcia, 2018).

Muir et al. (2019) comprehensively summarized PFASs in biota for terrestrial environments, freshwater systems, and marine environments in the Arctic. In general, PFOA, PFNA and PFDA predominated in vegetation, while PFNA, PFOS, PFUnDA, PFDA, perfluorotridecanoic acid (PFTrDA) and PFHxA were the main PFAS in wolf, moose and caribou liver in northern Canada and in reindeer and Arctic fox from Svalbard (Norwegian Environment, 2013). Precursors of PFCAs, such as 6:2 and 8:2 FTSs, were detected in the liver of Arctic fox from Svalbard. Generally, C9–C12 PFCAs predominate in freshwater fish in the Arctic (Ali et al., 2021a; Bossi et al., 2015; Bossi et al., 2005; Butt et al., 2010; Lescord et al., 2015; Muir et al., 2013; Norwegian Environment, 2013).

PFOS was the predominant PFAS in seals from Greenland and Svalbard. Relatively long odd-chain PFASs (PFUnDA, PFNA and PFTrDA) dominant PFCAs (Butt et al., 2014; Smythe et al., 2018). New PFASs have been reported in the liver of ringed seals from northwest Greenland and hooded seals from the Greenland Sea (Rotander et al., 2012), including perfluorotetradecanoic acid (PFTeDA), perfluoro-polyether carboxylate (PFPECA) and hentria-contafluoro- hexadecanoic acid (C16 PFCA) (Muir et al., 2015). 6:2 Chlorinated polyfluorinated ether sulfonate (6:2-Cl-PFAES) was detected in ringed seal liver from East Greenland, and perfluoroethylcyclohexane sulfonate (PFECHS) was found in ringed seal liver. Polyfluoroalkyl phosphate esters (mono- and diPAPs) in seal liver were below the MDL (<0.5 ng/g ww). PFOS, PFNA and PFUnDA were the dominant PFASs in polar bears from Greenland and Svalbard (Greaves et al., 2013; Boisvert et al., 2019).

Seabird eggs, liver and blood are typical matrices for monitoring PFAS levels in the Arctic (Butt et al., 2007; Lucia et al., 2015; Martin et al., 2004). PFUnDA and PFTrDA were the predominant PFCA in birds (Braune et al., 2014; Braune and Letcher, 2013; Vorkamp et al., 2019b). A recent survey showed that PFOS and long-chain PFCAs (>C8) were dominant in seabird eggs and marine mammals (Anna Kärrman et al., 2019).

2.9. Pharmaceuticals and personal care products (PPCPs)

PPCPs and their transformation products have been extensively measured in wastewater, river water and seawater in marginal seas (Kallenborn et al., 2018; Rodil et al., 2012; Tamura et al., 2017), while the transport of PPCPs via ocean currents to polar regions remains unclear (Arpin-Pont et al., 2016). AMAP (2017) conducted a comprehensive survey of the occurrence and fate of PPCPs in the Arctic, which raised concern for PPCPs in polar regions.

Among PPCPs, limited data are available for organic ultraviolet filter chemicals (UV-Fs) and synthetic fragrance materials (FMs) in the air. Galaxolide (HHCB) and tonalide (AHTN) were the two most abundant FMs in the air and have been detected in ship-bound air samplers from the European continent to the high Arctic, suggesting LRAT potential (Xie et al., 2007). Atmospheric UV-Fs emerged following their detection in both gaseous and particle phases in urban air in Canada (Pegoraro et al., 2020). In addition, UV-Fs were found in streams formed by melting snow and ice in the Antarctica.

As a typical volatile compounds, cyclic volatile methylsiloxanes (cVMSs) were first reported in the air collected at Zeppelin station in 2011 (Krogseth et al., 2013). In 2020, the average concentrations of octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5) and dodecamethylcyclohexasiloxane (D6) in the air were 0.5, 1.04 and 0.21 ng/m^3 at Zeppelin and 1.29, 4.87 and 0.46 ng/m^3 at Birkenis, respectively (Bohlin-Nizzetto et al., 2021). No clear temporal trends were observed. More studies on UV-Fs and FMs in the air from polar regions are required to reveal their LRAT. D4 and D5 were the predominant compounds, with concentrations from MDLs to 110 ng/g dw in the soil (Sanchis et al., 2015).

In aquatic environments, >50 PPCPs were found in both the influent and effluent of sewage, with concentrations from a few ng/L to μg/L resulting from the Nordic environmental screening program (Kallenborn et al., 2018; Schlabach et al., 2017). Metabolites and transformation products of PPCPs were also identified in sewage effluent (Fig. 9) (Kallenborn et al., 2018; Weigel et al., 2004). Stroski et al. (2020) reported 7 PPCPs (atenolol, carbamazepine, metoprolol, naproxen, sulfapyridine, sulfamethoxazole, and trimethoprim) in wastewater effluents from four Canadian Arctic communities (Fig. 9).

FMs such as HHCB and AHTN were found in seawater from the European Arctic at mean concentrations of 59 and 23 pg/L, respectively (Xie et al., 2007). Tsui et al. (2014) reported that benzophenone-3 (BP-3), 4-methoxycinnamate (EHMC) and octocrylene (OC) were dominant UV-Fs in seawater from the Arctic at concentrations of 16.6, 25.4 and 25.8 ng/L. FMs were measured in seawater and surface snow up to 72 ng/L in Ny-Ålesund Svalbard (Vecchiato et al., 2018). Generally, the levels of UV-Fs in the Arctic were 1–2 orders of magnitude lower than those from urban environments (Tsui et al., 2014).

A range of PPCPs have been determined in the wastewater effluent of research bases in Antarctica (Emnet et al., 2015; Esteban et al., 2016). The concentrations of methyl-triclosan, 4-methyl-benzylidene camphor, octylphenol and benzophenone-1 (BP-1) were comparable to the levels in the urban environment. Sewage discharge and disposal of raw human waste contributed major PPCPs in the Antarctic coastal environment. Alkylphenol (1.14–7225 ng/L) was measured in the surface water, including streams, ponds, glacier drains and seawater, in the Northern Antarctic

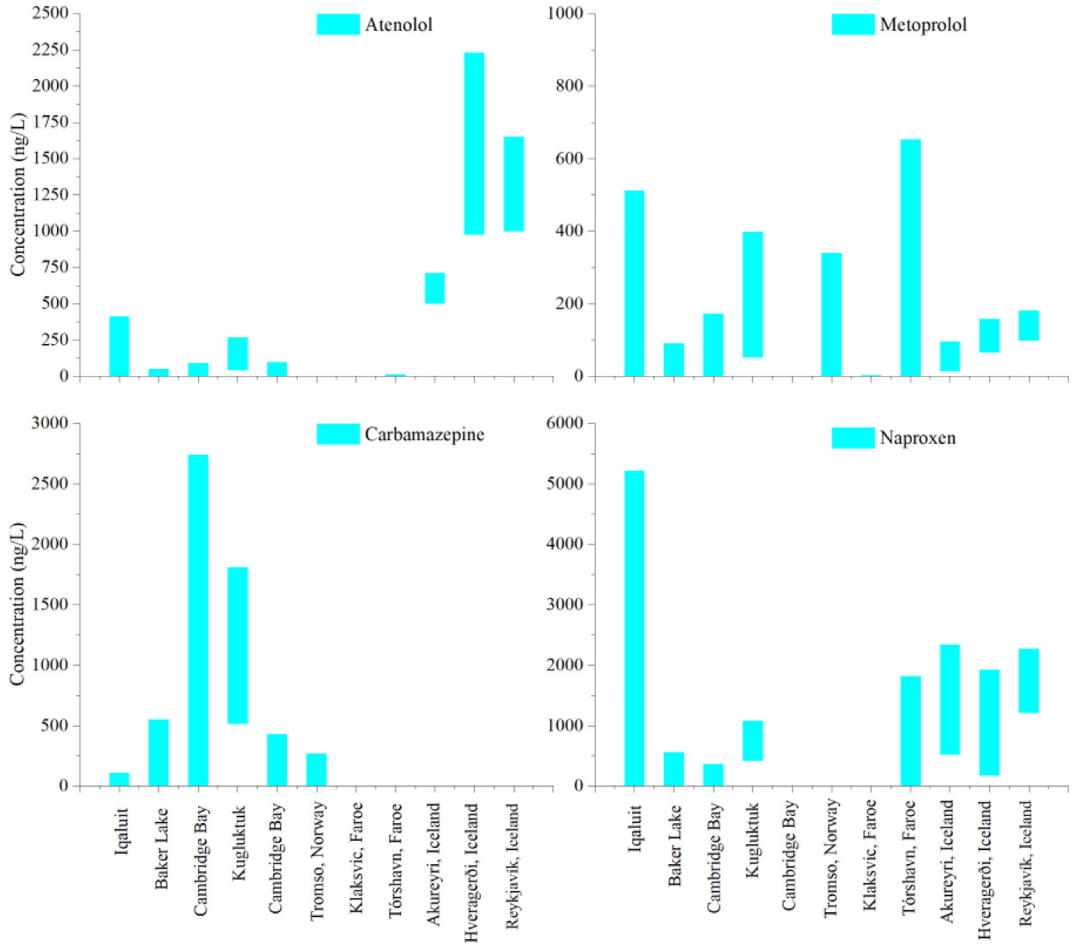

Fig. 9. Concentration ranges (ng/L) of atenolol, metoprolol, carbamazepine and naproxen in effluent waters in the Arctic (Chaves-Barquero et al., 2016; Huber et al., 2016; Stroski et al., 2020; Weigel et al., 2004).

Peninsula region. UV-Fs were found in both water and particle phases (Dominguez-Morueco et al., 2021).

Acetaminophen (48.74 μg/L), diclofenac (15.09 μg/L), ibuprofen (10.05 μg/L) and stimulant caffeine (71.33 μg/L) were the dominant PPCPs in waters discharged directly into the ocean without any prior purification processes (Gonzalez-Alonso et al., 2017). Eight antibiotics were determined in treated wastewater from King George Island with concentrations ranging from 0.001 (trimethoprim) to 0.89 μg/L (quinolones ciprofloxacin) (Hernandez et al., 2019). Ciprofloxacin (4–218 ng/L) was also detected in seawater, suggesting discharge from wastewater to the coastal environment. FMs were detected in seawater of Terra Nova Bay, with total concentrations up to 100 ng/L (Vecchiato et al., 2017). Apart from some local sources, the increasing concentrations measured in the seasonal meltwater implied reemissions of FMs from sea ice and snow in Antarctica.

Warner et al. (2010) investigated cVMS in sediment and biota in the European Arctic. The concentrations of D5 were 60 ± 1.2 in Chironomidae larvae, 107 ± 4.5 in pea clams Pisidium sp., 131 ± 105 in sticklebacks, 41 ± 38 in char, and 9.9 ± 5.9 (trout) ng/g ww from Lake Storvannet in Northern Norway. Relatively high levels of D5 were measured in Atlantic cod (176 ng/g lw) and shorthorn sculpin (531 ng/g lw). VMSs have also been determined in vegetation, phytoplankton, and krill from the Antarctic Peninsula region (Sanchis et al., 2015). UV-Fs, parabens and alkylphenol were detected in biota from coastal areas near the Antarctic research stations (Emnet et al., 2015). There are very few data for pharmaceuticals in biota. Given their high concentrations in effluents and coastal water, more studies are needed to determine the bioaccumulation of certain PPCPs in marine fish and mammals.

3. Sources, long-range transport and environmental pathways

3.1. Sources of POP and EOCs

POPs and EOCs can be released to the environment from direct and indirect sources. Halogenic flame retardants and plastic-related chemicals such as BFRs, OPEs, and UV-Fs are usually physically added to the products. They can leach out during the production, application and disposal of industrial and household products (Alcock et al., 2003). E-waste recycling is an important point source for BFRs, OPEs, plasticizers and UV-Fs (Perkins et al., 2014). Other point sources of BFRs and OPEs in the environment are municipal or hazardous waste incinerators and leachates from waste disposal and effluents from factories producing flame-retardant polymers (Clarke et al., 2008; Zhang et al., 2019).

PAHs and their derivatives are derived from primary combustion sources as well as atmospheric secondary formations (Atkinson and Arey, 1994). As with the source apportionment results in multimedia from low-latitude areas (Albinet et al., 2008; Alves et al., 2017), nitrated, hydroxylated, and oxygenated PAHs were primarily sourced from combustion-related emitters based on few available reports in polar regions (Drotikova et al., 2020).

Photodegradation and reaction with OH, Cl or O_3 radicals are the major pathways to eliminate neutral PFAS from the atmosphere In polar regions (Ellis et al., 2004; Ellis et al., 2003; Loewen et al., 2005), especially in spring and summer. Degradation of fluorotelomer alcohol (FTOH) fluorinated sulfonamides (FOSAs) and fluorinated sulfonamidoethanols (FOSEs) in the aquatic phase can be an additional source for perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane sulfonic acids (PFSAs), especially in remote oceans (Armitage et al., 2009; Cousins et al., 2011). In addition,

increasing human activities in the Arctic and the Antarctica may directly release PFASs into the polar environments (Wild et al., 2015).

Inadequate wastewater treatment facilities at the research stations and settlements could result in the direct release of untreated or undertreated wastewater to the coastal environment (Gunnarsdottir et al., 2013). Thus, wastewater runoff could be an important local contamination source of PPCPs.

3.2. Long-range environmental transport (LRET)

EOCs and POPs can undergo LRAT after volatilization from their sources (Fig. 10) (Lakaschus et al., 2002; MacDonald et al., 2000; Wania and Dugani, 2003). Apart from LRAT, the transport of EOCs and POPs associated with ocean currents was investigated in pan-Arctic shelf seas and the interior basin, resulting in the emissions of PBDEs and OPEs in a minor fraction being ultimately transported long-range to the Arctic (Li et al., 2017b; Na et al., 2020b; Salvadó et al., 2016). Recent studies on OPEs in both the Arctic and Antarctica provide evidence that OPEs, especially chlorinated OPEs, can be efficiently transported via ocean currents and the atmosphere from source areas to polar regions (Rodgers et al., 2018; Suhring et al., 2021; Suhring et al., 2016). Long-range transport of atmospheric PAHs and derivatives has been evidenced by continuous monitoring from remote sites in the Arctic and/or modeling simulation (Hung et al., 2005).

The presence of OCPs, PCBs, PCNs, OPEs, PFASs and CPs in polar areas has been attributed to cold condensation and global fractionation during LRAT (Frank, 2003). Therefore, atmospheric transport is the primary pathway in winter (Macdonald et al., 2005). Moreover, both observations and model predictions have shown that OCPs and PCBs can be transported from low-latitude zones to polar regions by multiple transfers, i.e., repeated cycles of deposition and re-evaporation (Lakaschus et al., 2002; Lohmann et al., 2001; MacDonald et al., 2000). In addition, ocean currents and river inputs played an important role in the entry of OCPs, PCBs, OPEs and PPCPs into the Arctic Ocean (Li et al., 2002; Zhulidov et al., 2000).

Owing to their unique properties, PFAS can be transported from sources to polar regions via both atmospheric and oceanic currents (Muir and Miaz, 2021; Xie et al., 2015). FTOH, methyl and ethyl perfluorooctane sulfonamides (MeFOSA and EtFOSA); and methyl and ethyl perfluorooctane sulfonamidoethanols (MeFOSE and EtFOSE) mainly travel with air masses from source regions to remote areas (Lin et al., 2020a; Taniyasu et al., 2013; Wang et al., 2015). Ionic PFASs mostly accumulate in surface waters, and their LRET is governed by global ocean circulation (Ahrens et al., 2010; Nash et al., 2010; Yamashita et al., 2008; Yamazaki et al., 2021). Marine aerosols or sea spray have been proven to be important media to transport ionic PFASs over the ocean surface and contribute to LRAT and deposition in the Arctic and Antarctica (Casas et al., 2020; Maclnnis et al., 2019; Wolf et al., 2021).

Although some PPCP compounds were detected in the polar regions, the long-range transport potential of the PPCP substances still remain to be elucidated. Moreover, high concentrations of PPCPs measured in the coastal waters in the polar regions are not necessarily related to higher consumption rates but might result from the higher environmental stability under the low-temperature climate in the Arctic and Antarctica. Volatile and semivolatile PPCPs can undergo LRAT.

3.3. Environmental pathways

Air-sea gas exchange fluxes of EOC and POPs have been studied previously in polar oceans (Möller et al., 2011a & b). Cabrerizo et al. (2019) showed that PCBs and OCPs shifted from equilibrium during ice/snow-covered conditions toward a clear net volatilization of PCBs and most of the OCPs during snow/ice-free conditions in the Arctic rivers, lakes and oceans (Fig. 10). PAH outgassing and absorption into seawater were found in the Arctic Ocean (Ma et al., 2013; Zheng et al., 2021). With respect to air-snow exchange for PAHs in the Antarctica, both a near equilibrium state and net volatilization exist (Cabrerizo et al., 2014; Casal et al., 2018). In the Southern Ocean between Australia and Antarctica, estimated air-seawater fluxes showed net deposition between 200 and 6400 pg/m^2/day OCPs (Bigot et al., 2016). Positive volatilization fluxes for PCBs, HCB, heptachlor and DDTs were shown along the Western Antarctic Peninsula (Khairy et al., 2016). TCIPP and TCEP exhibited net deposition fluxes from the North Pacific to the Arctic, while net volatilization fluxes dominated in the European Arctic (Li et al., 2017b; Na et al., 2020b). Net volatilization of FTOH from snow and ocean to air has been demonstrated in Antarctica (Wang et al., 2015) and Ny-Ålesund (Xie et al., 2015). The deposition fluxes were 1.5 and 5.7 ng/m^2/day for HHCB and AHTN in the European Arctic (Xie et al., 2007).

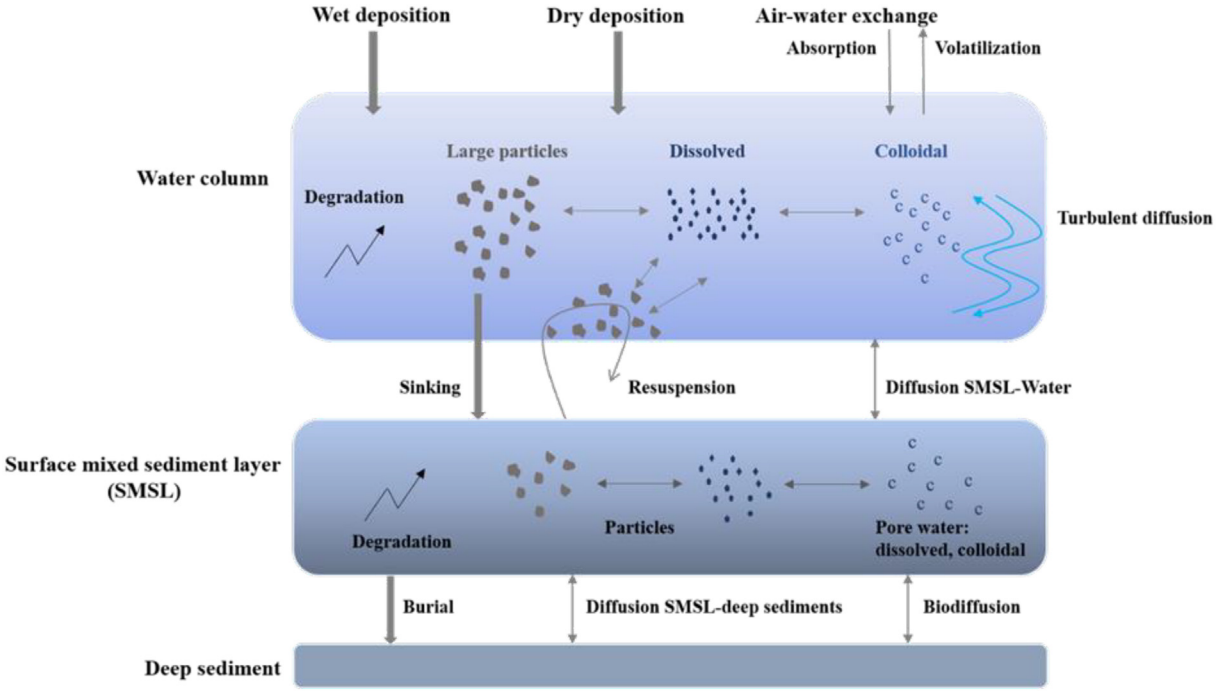

Fig. 10. Biogeochemical cycling of organic contaminants in the marine environment (Lohmann et al., 2007).

In polar regions, snow deposition acts as an effective scavenger for atmospheric EOCs and POPs in the Arctic, Southern Ocean and Antarctica (Fig. 10) (Cheng et al., 2013b; Li et al., 2017a, 2017b; Xie et al., 2020). Along with the processes of ice retreat and snow melting, POPs trapped in snow could be released into the water column and atmosphere. Consequently, elevated PBDE, OPE, PFAS and OCP concentrations were measured in the high Arctic Lake rather than in the North Atlantic Ocean and the Northeast Pacific Ocean (Li et al., 2017a, 2017b; Na et al., 2020a, 2020b; Sun et al., 2020).

3.4. Bioaccumulation

Studies have demonstrated the biotransformation of higher brominated PBDEs to lower brominated congeners, generally correlated with high levels of bioaccumulation (Riu et al., 2008). Both trophic magnification factors and species-specific bioaccumulation factors of PBDEs were analyzed in the Canadian Arctic marine food web. It is suggested that PBDEs are absorbed by homeotherms and accumulate rather than biomagnification, exhibiting a relatively rapid depuration rate though biotransformation in Arctic marine organisms (Kelly et al., 2008). Similar bioaccumulation but limited biomagnification potential was recorded in the terrestrial food chain of the Canadian Arctic (Morris et al., 2018) as well as PBDEs in the food web of the Ross Sea, Antarctica (Corsolini et al., 2017). The trophic magnification factors indicated that the levels of p,p-DDE, p,p-DDD, cis-chlordane, and β-HCH were magnified through the food web (Kim et al., 2021). PAHs with sufficient lipophilicity could be subject to trophic transfer of PAHs in marine food webs, revealing an inverse correlation between PAH burdens and trophic position and therefore suggesting that biomagnification in food webs is insignificant (Perugini et al., 2007). Despite the existence of PAH bioaccumulation in invertebrates, fish, and seabirds in polar regions (Perugini et al., 2007), this is not true for marine mammals due to the trophic dilution effect. Seal blubber to bear liver biomagnification factors (BMFs) reflected the dietary exposure relationship of PFAS between bears and seals (Boisvert et al., 2019). The BMFs of C9–C13 PFCAs decreased with increasing chain length. In a marine food web from the coastal area of Longyearbyen (Ali et al., 2021a), PFAS profiles changed with increasing trophic level, with high percentages of 6:2 FTS, FOSA and long-chain PFCAs in zooplankton and polychaetes, while linear PFOS dominated in fish and gull liver. Moreover, 6:2 FTS and PFBS showed bioaccumulation potential in marine invertebrates, whereas they were not detected in organisms at higher trophic levels.

3.5. Impact of climate change

Polar regions have been experiencing strong warming processes rather than other parts of the earth, which has induced changes in the emission sources, transport and biogeochemical cycle of organic contaminants in the Arctic and Antarctica, ultimately affecting human exposure. Moreover, an assessment of the interaction between Arctic climate change and POPs was carried out by AMAP in 2019–2021 under unprecedented changes in Arctic snow, water, ice and permafrost conditions (AMAP, 2021). Nadal et al. reviewed the climate change impact on the concentrations of POPs, and an intensification of studies to identify and mitigate the indirect effects of climate change on POP's fate was required to minimize the human health impact (Nadal et al., 2015). Climate change is considered to enhance POPs LRET, and remote areas such as the Arctic and Antarctica are considered likely to be the most impacted under the climate change perspective (Teran et al., 2012). Monitoring programs oriented toward the observation of secondary POP's sources and the enhancement of inventories reporting primary and secondary POPs emissions are useful in dealing with POPs exposure under climate change scenarios.

4. Conclusions and perspectives

Legacy POPs in the environmental media and biota have shown declining temporal trends in the Arctic and the Antarctica in accordance with the global attempt to eliminate their manufacture and application. However, previously released legacy POPs are likely entering the global cycle again due to climate change. Thus, continuous monitoring of the temporal variability in legacy POPs in environmental matrices is needed.

EOCs are widely present in various environmental matrices and organisms in the Arctic and Antarctica. Studies in the Arctic have shown that EOCs are the main chemical groups in both the environment and biota and exceed the levels of legacy POPs. Both atmospheric and oceanic currents can carry EOCs into the polar regions. Moreover, enriched human activities such as shipping, research expeditions and tourism in the polar regions can directly contribute to the local environment. Therefore, background measurements of EOCs need to be considered to minimize interference. Current research has mostly focused on oceanic transects and aquatic environments. Comprehensive surveys and long-term monitoring of EOCs in the air, water and terrestrial biota in the Arctic and Antarctica need to be strengthened to elucidate their LRET, persistence, bioaccumulation and toxicity (Fu et al., 2021; Xie et al., 2022).

The transportation pathways of EOCs to the Arctic and the Antarctica are not well understood and suffer from a paucity of observations, both spatially and temporally. Many models have been established to predict the fate and transport of POPs from source regions to polar areas and attempt to simulate and illuminate the environmental pathways for EOCs. However, the accuracy of these models depends on the resolution of the geographic map, the density of meteorological observations, physical/chemical processes such as gas-particle partitioning, and photo/biological degradation. In addition, the flux of EOCs with terrestrial and fresh water (rivers, melting ice and snow) input to the Southern Ocean and Arctic, sedimentation and resuspension require intensive study. Future modeling efforts need to incorporate more accurate properties of EOCs, such as bioaccumulation factors, particle deposition, and air-water/snow exchange fluxes. In addition, accurate and high-resolution EOC data are required to improve model predictions for their environmental fates and health impact.

The establishment of concentration thresholds for individual compounds as well as for realistic cocktail mixtures that in fact indicate biologically relevant and not statistically determined. It is necessary to establish and refine concentration thresholds for biologically relevant health effects in wildlife and fish of polar areas. Moreover, the multiple stressors of POPs and EOCs under changing climate should be explored to assess the actual health risks of humans and animals in the polar regions (Dietz et al., 2019; Szopinska et al., 2019).

Geographic variation for both legacy POPs and EOCs among monitoring stations mainly reflected the different origins of emission sources. Unexpected high or low concentrations have sometimes been reported in the polar regions, especially for the levels of EOCs. The uncertainties could be addressed to not only the background contamination but also different sampling techniques and operational procedures. Furthermore, evaluations of the environmental occurrences of EOCs in the polar regions are highly limited by diverse sampling and analytical strategies applied by different research groups. Therefore, systematic monitoring programs for EOCs in the polar regions need to be performed urgently by national and international organizations, such as AMAP and SCAR.

Climate change directly and indirectly impacts the sources, transport pathways, and environmental fate of POPs and EOCs (AMAP, 2021). Reemission of legacy POPs and EOCs accumulated in glaciers, sea ice, and snow may alter the concentrations and amplify their effects in the polar regions. Thus, future research will need to understand the various biogeochemical and geophysical processes under climate change and anthropogenic pressures to be able to predict the environmental fates and toxicity risk of EOCs in the polar regions.

CRediT authorship contribution statement

ZX: Conceptualization, Writing-original draft, Review & editing; Project coordination. PZ: Writing-original draft, Review & editing. ZW: Writing-original draft, Review & editing. SZ: Writing-original draft, Review & editing. LW: Writing-original draft, Review & editing. LM: Data analysis,

Writing-original draft, Review & editing. AK: Comment, Review & editing. JG: Comment, Review & editing. RE: Review & editing. RY: Writing-original draft, Review & editing. ZW: Writing-original draft, Review & editing, WM: Writing-original draft, Review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests.

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Appendix A. Supplementary data

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