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Final report

Emerging and legacy organic contaminants in the polar regions

by:

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Abstract: Emerging and legacy organic contaminants in the polar regions - Final Report

Organic contaminants in polar regions have become significant concerns because of their persistence, bioaccumulation and toxicity potential. Climate change can alter the biogeochemical cycling of persistent organic pollutants (POPs) and emerging organic contaminants (EOCs) and amplify their effects on polar ecosystems. Occurrences of POPs and EOCs from long-range transport and local discharge have left impacts on fragile polar ecosystems. Therefore, actions are urgently needed to monitor the temporal trends of POPs and to investigate novel EOCs in polar regions. The data on legacy POPs in environmental media and biota exhibit declining trends in both the Arctic and the Antarctic by virtue of the global endeavor in banning their manufacture and usage. However, the reemission of POPs that previously accumulated in the polar environment has been observed, and these POPs can enter the global cycle again following the processes of ice retreat, glacier melting and permafrost thawing driven by global warming. Therefore, continual monitoring should be conducted for legacy POPs in polar areas. Screening surveys for EOCs in environmental and biological matrices have been carried out through national and regional research programs. The long-range environmental transport of EOCs has been highlighted with their occurrences in ice cores, snow, and lake waters in polar regions. Therefore, the investigation of EOCs in the Antarctic needs to be strengthened through national and international research programs. Glacial ice and snow acted as secondary emission sources in the polar regions and released POPs and EOCs into the atmosphere and ocean. 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 polar regions.

Kurzbeschreibung: Neuartige und klassische organische Schadstoffe in den Polarregionen – Abschlussbericht

Organische Schadstoffe in Polarregionen sind aufgrund ihrer Persistenz, Bioakkumulation und ihres Toxizitätspotenzials zu erheblichen Bedenken geworden. Der Klimawandel kann den biogeochemischen Kreislauf von persistenten organischen Schadstoffen (POPs) und neuartigen organischen Schadstoffen (EOCs) verändern und ihre Auswirkungen auf polare Ökosysteme verstärken. Das Auftreten von POPs und EOCs durch Ferntransport und lokalen Austritt hat Auswirkungen auf empfindliche polare Ökosysteme hinterlassen. Daher sind dringend Maßnahmen erforderlich, um die zeitlichen Trends von POPs zu überwachen und neue EOCs in Polarregionen zu untersuchen. Die Daten zu klassischen POPs in Umweltmedien und Biota zeigen aufgrund der weltweiten Bestrebungen, ihre Herstellung und Verwendung zu verbieten, sowohl in der Arktis als auch in der Antarktis rückläufige Trends. Es wurde jedoch die Reemission von POPs beobachtet, die sich zuvor in der polaren Umgebung angesammelt haben, und diese POPs können nach den durch die globale Erwärmung verursachten Prozessen des Eisrückgangs, des Gletscherschmelzens und des Permafrostauftauens wieder in den globalen Kreislauf eintreten. Daher sollte in Polargebieten eine kontinuierliche Überwachung von klassischen POPs durchgeführt werden. Screening-Erhebungen auf EOCs in Umwelt- und biologischen Matrizes wurden durch nationale und regionale Forschungsprogramme durchgeführt. Der weiträumige Umwelttransport von EOCs wurde durch ihr Vorkommen in Eisbohrkernen, Schnee und Seewasser in Polarregionen hervorgehoben. Daher muss die Untersuchung von EOCs in der Antarktis durch nationale und internationale Forschungsprogramme verstärkt werden. Gletschereis und -schnee fungierten als sekundäre Emissionsquellen in den Polarregionen und setzten POPs und EOCs in Atmosphäre und Ozean frei. Daher muss die zukünftige Forschung die verschiedenen biogeochemischen und geophysikalischen Prozesse unter Klimawandel und anthropogenen Belastungen verstehen, um das Umweltverhalten und das Toxizitätsrisiko von EOCs in Polarregionen vorhersagen zu können.

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List of abbreviations

APP	Arctic pollution potential
АМАР	Arctic Monitoring and Assessment Programme
BAF	Bioaccumulation Factor
BFR	Brominated flame retardant
СР	Chlorinated paraffin
DDT	Dichlorodiphenyltrichloroethane
dl-PCBs	dioxin-like PCBs
EOC	Emerging organic contaminant
EPA	Environmental Protection Agency
FR	Fugacity ratio
GAPS	Global Atmospheric Passive Sampling
ImPACT	Input Pathways of persistent organic pollutants to Antarctic
K _{AW}	Air-water partition coefficient
ΚοΑ	Octanol-air partition coefficient
Kow	Octanol-water partition coefficient
LCCPs	Long-chain chlorinated paraffins
LRAT	Long-range atmospheric transport
LRET	Long-range environmental transport
LRTAP	Long-range transboundary air pollution
MCCPs	Medium-chain chlorinated paraffins
OCPs	Organochlorine Pesticides
OPEs	Organophosphate esters
PAHs	Polycyclic aromatic hydrocarbons
PBDEs	Polybrominated diphenyl ethers
PCBs	Polychlorinated biphenyls
PCDDs	Polychlorinated dibenzo-p-dioxins
PCDFs	Polychlorinated dibenzofurans
PCN	Polychlorinated naphthalene
PFASs	Per- and poly fluoroalkyl substances
POPs	Persistent and organic pollutants
PPCPs	Pharmaceuticals and personal care products
SC	Stockholm Convention
SCAR	Scientific Committee on Antarctic Research
SCCPs	Short-chain chlorinated paraffins
TMFs	Trophic Magnification Factors
UNECE	United Nations Economic Commission for Europe
UNFCCC	United Nations Framework Convention on Climate Change

Summary

Legacy (persistent) and emerging organic contaminants (POPs and EOCs) in polar regions have become significant concerns because of their persistence, bioaccumulation and toxicity potential. Climate change can alter the biogeochemical cycling of POPs and EOCs and amplify their effects on polar ecosystems. Global industrialization and reconstruction have affected both the Arctic and the Antarctic, which has led to an increase in human activities such as tourism, shipping, resource exploration and the construction of research stations. Occurrences of POPs and EOCs from long-range transport and local discharge have left impacts on fragile polar ecosystems. Therefore, actions are urgently needed to monitor the temporal trends of POPs and to investigate novel EOCs in polar regions.

The Arctic Monitoring and Assessment Programme (AMAP) was established in1991 to implement components of the Arctic Environmental Protection Strategy (AEPS) and has become a program group of the Arctic Council. The AMAP has launched several research programs for monitoring classic persistent organic pollutants (POPs) and emerging organic pollutants (EOCs) in air, sediment, and organisms in the pan Arctic. In recent decades, the AMAP has published periodic assessments for classic contaminants and emerging chemicals that may be of Arctic concern and the climate change effects. Scientific information has been transferred to support stakeholders of Arctic governments in taking remedial and preventive actions related to contaminants. For the Antarctic, the Scientific Committee on Antarctic Research (SCAR) has a long and successful record of summarizing policy-relevant scientific knowledge to policy makers through its groups such as the Input Pathways of persistent organic pollutants to Antarctic (ImPACT). The experiences of the AMAP have been successfully transferred for the observation and assessment of chemical contaminants in the Antarctic through the research projects and activities of ImPACT.

The data on legacy POPs in environmental media and biota exhibit declining trends in both the Arctic and the Antarctic by virtue of the global endeavor in banning their manufacture and usage. However, the reemission of POPs that previously accumulated in the polar environment has been observed, and these POPs can enter the global cycle again following the processes of ice retreat, glacier melting and permafrost thawing driven by global warming. Therefore, continual monitoring should be conducted for legacy POPs in polar areas. However, a number of EOCs have been reported in different environmental matrices in the Arctic and Antarctic. Some EOCs, such as per- and poly fluoroalkyl substances (PFASs), chlorinated paraffins (SCCPs and MCCPs) and organophosphate esters (OPEs), have been included in long-term atmospheric monitoring programs of the AMAP. Screening surveys for EOCs in environmental and biological matrices have been carried out through national and regional research programs. Data on EOCs in Antarctic are rare. Nevertheless, the long-range environmental transport of EOCs has been highlighted with their occurrences in ice cores, snow, and lake waters in polar regions. Therefore, the investigation of EOCs in the Antarctic needs to be strengthened through national and international research programs.

Environmental sample banks (ESBs) for polar species have been established with support from national research programs, especially in the Arctic states. Having these banks provides the best chance to reconstruct the historic fingerprint of chemical contaminants recorded in the polar environment. However, the ESB for the Antarctic is quite limited. Therefore, a long-term monitoring program in the Antarctic should be initiated to collect typical environmental and biological samples to establish an Antarctic ESB.

An assessment of the interaction between Arctic climate change and POPs and chemicals of emerging Arctic concern was carried out by the AMAP in 2019-2021 under unique changes in

Arctic snow, water, ice and permafrost conditions. Legacy POPs, such as organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs), showed a strong response to specific climate change scenarios. Glacial ice and snow acted as secondary emission sources in the polar regions and released POPs and EOCs into the atmosphere and ocean. 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 polar regions.

Polybrominated diphenyl ethers (PBDEs)

Despite application controls on the penta- and octa-PBDE products beginning to result in declines in concentration in environmental samples, there is still an urgency to conduct a continuous survey of the temporal trends of PBDEs from the polar environment in cases where the PBDE pollution status is uncertain. More data on the temporal variability of PBDE levels and distributions in environmental matrices and sampling sites are therefore needed. PBDEs are generated on land by human activities, with secondary emissions from contaminated reservoirs. However, reports on PBDE air-surface exchange in the Arctic/Antarctic are scarce, impeding our understanding of whether previous "sinks" transition into a source. In the context of climate change, the impact of global warming on PBDE transport and fate in polar areas as well as the temporal tendency and bioaccumulation in biota are largely unexplored. Considering some evidence of decabromodiphenyl ether (BDE-209) enrichment in Antarctic biota, the long-range transport and deposition of BDE-209 to the Antarctic deserves more attention. Since novel brominated flame retardants (BFRs) are being marketed for PBDE replacement, investigation of the occurrence and emission trends of these burgeoning alternatives in polar environments is recommended.

Polycyclic aromatic hydrocarbons (PAHs)

Broad-scale spatial analysis for PAHs and particularly their derivatives from terrestrial and marine environments in polar regions remains lacking. Recent studies of the temporal trends of PAHs from air monitoring and dated cores are limited. Polar PAHs such as oxygenated and nitrated compounds possess more genotoxic, mutagenic, and carcinogenic effects than their parent PAHs. However, environmental and human health implications as well as assessments of the relative primary and secondary inputs of PAH derivatives in polar regions have been largely unexplored. Several PAHs are converted into long-lived derivatives after the photochemistry process and can undergo long-range atmospheric transport (LRAT). The assessment of LRAT potential of PAH derivatives, in particular nitro-PAHs, should be extended. The modulation of atmospherically transported PCBs by oceanic biological pumps in the Arctic has been reported. However, against the backdrop of possibly declining PAH emissions from primary sources in the long run as well as global warming, the biological pump effect may be attenuated due to the increasing release of PAHs into air from local secondary sources and the migration of longpreserved PAHs from sea ice into water as a result of enhanced ice melting. The coupling between the environmental fate of PAHs in polar regions and climate change or anthropogenic activity deserves long-term attention.

Organochlorine pesticides (OCPs)

The lipid contents and tissue specific protein of the organisms and other biochemical processes controls the bioaccumulation and transformation processes of OCPs. An enhanced, selective retention of certain organohalogen classes in given tissues/body compartments may thus lead to site-specific toxicological actions and adverse effects in highly contaminated Arctic birds. Further investigation of the mechanisms underlying macromolecular binding interactions and organohalogen accumulation in birds is required (Verreault et al., 2007). As legacy OCPs such as

dichlorodiphenyltrichloroethane (DDT) was phased out since 1970s, many new pesticides have been applied as replacements to serve similar purposes. In theory, current-use pesticides (CUPs) have been licensed for use owing to their low persistence and bioaccumulation potentials. Despite their lower concentrations in biota of the Arctic compared to legacy OCPs, biomodification effects have been found, and their long-range transport (LRT) potential has been proven as well. Therefore, continued monitoring of both OCPs and CUPs in polar regions are necessary for future research programs.

Polychlorinated biphenyls (PCBs)

It is clear that PCBs in environmental media and biota exhibit a declining tendency both in the Arctic and in the Antarctic by virtue of the global endeavor in banning PCB manufacture and use. However, PCBs previously deposited in the environment are likely to enter the global cycle again due to climate change. Therefore, we should continue to pay close attention to PCB legacy in polar areas. We have reviewed a number of existing studies and summarized several knowledge gaps among scientific research, decision making and management policies. The unintentionally produced (UP) PCBs, such as PCB-5 and PCB-11, has caused increasing concern (Bartlett et al., 2019). PCB-11 was determined in the surface snow collected from Lomonosovfonna, Svalbard, in 2009-2010 and 2013-2014, accounting for 4% of the total PCBs (Garmash et al., 2013). PCB-11 and other UP-PCBs are not well understood in the Arctic. Although their concentrations were not as high as those of some legacy PCBs, the toxicity of the parent source and metabolites is still unclear, which might cause a higher negative risk effect.

In most previous research, approximately 2 or 3 decades of PCB congeners were targeted, accounting for <10% of total PCBs. Other UP-PCBs in polar areas are less understood, such as PCB-11, which contributed to 8.5% of total PCBs in more recent studies (Bartlett et al., 2019); thus, more measurements of all 209 PCB congeners are essential if we are to begin to understand their contributions to environmental contamination. In addition, for future risk assessment, we recommend exploring the synergistic toxic effects of multiple contaminants and filling the gaps in spatial distribution.

The transportation pathways of POPs to the Arctic and the Antarctic are not well understood because of limited observations, both spatially and temporally. Many models have been established to predict the fate and transport of PCBs from source regions to polar areas. However, the accuracy of these models depends on the resolution of the geographic map, the density of meteorological observations, and the physical/chemical processes (cloud, fog, precipitation, temperature and sunlight, as well as air-sea exchange and gas-particle exchange, etc), whose influences are inadequately assessed in the models. With respect to the terrestrial/freshwater pathways of POPs into the Arctic, research on the air-snowpack exchange, flux of terrestrial and fresh water input into the ocean, sediment resuspension contributions and ice cover influence has been poorly understood. In addition, ocean and ice circulation models used for contaminant transport often have low resolution that can not resolve boundary currents and eddies. Furthermore, contaminants enter the models only as passive tracers. Therefore, future modeling efforts need to incorporate more realistic properties, including bioaccumulation, particle scavenging and degradation rates.

Concentration thresholds need to be determined for both individual compounds and cocktail mixtures that demonstrate biological relevance but have not been statistically documented. On the one hand, concentration thresholds should be established for biologically relevant health effects in wildlife and fish in polar areas. On the other hand, identification of cumulative and interactive effect thresholds of contaminant exposure need to be strengthen. The multiple stressors of chemical contaminants and climate change should be explored to assess the actual

health risks to human being and organisms (Dietz et al., 2019; Letcher et al., 2010; Szopińska et al., 2019).

Chlorinated paraffins (CPs)

At present, research on the environmental behavior of short-chain (C10-C13) chlorinated paraffins (SCCPs) is largely limited by their quantitative analysis, and the existing analysis methods cannot screen and analyze SCCP monomers individually. In terms of the selection of standard products, most laboratories mainly take the commercial industrial product mixture as the standard. However, most SCCPs released into the environment undergo selective environmental migration and biological metabolic transformation, and their composition may be quite different from that of commercial mixtures. This difference is finally reflected in the inconsistency of the characteristics of SCCPs between the analytical standard and the actual sample, which is then transmitted to the difference of the response of the instrument detector to different laboratories, analytical standards, pretreatment and instrumental analysis methods in the analysis process, it is difficult for to compare SCCPs horizontally among studies of ecosystem occurrence and environmental behavior. Therefore, it is urgent to develop suitable analytical methods with higher sensitivity and accuracy.

Polychlorinated Naphthalenes (PCNs)

Although the occurrences of PCNs have been detected in various environmental matrices, including air, biota and mammals, there has been a lack of systematic monitoring projects to study the temporal diversification of PCNs and evaluate the implementation effect of the Stockholm Convention. To our knowledge, the sources of PCNs mainly include long-distance atmospheric migration and anthropogenic emission sources, but the proportion of each type is not clear. Hence, a cleaner background point should be selected for the specific sampling site for investigation, and even the anthropogenic emissions generated by the monitoring station itself should be considered.

In addition, we do not know the toxic changes in PCN degradation products in special polar environments, so it is necessary to analyze, monitor and comprehensively characterize the degradation products of PCNs. Previous studies investigated few polar biological species and lacked systematic comprehensive research on various species in the structure of the food chain, so biomagnification of PCNs in the food chain in polar regions is worthy of further study. In today's global warming scenario, the melting of ice and snow leads to the incremental release of stored POPs. Therefore, it is necessary to continuously observe the biogeochemical and geophysical processes of these POPs (including PCNs) to predict their environmental fate and toxicological risks.

Dioxin

The detection in these remote areas shows that they can continuously accumulate in polar areas, especially for polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), which can be stably retained in these areas. Therefore, it is necessary to pay attention to the temporal change trend of these pollutants. Through systematic research and monitoring, we can continuously pay attention to these pollutants and establish a model to predict the future environmental fate. As a byproduct of incomplete combustion reactions and chemical production, dioxins are still produced continuously, but the scale of human use of combustion reactions is rapidly growing; furthermore, human demand for chemical products is increasing rapidly, so the generation of dioxins cannot be avoided. Global warming is continuously causing the melting of ice and snow in Antarctic and the Arctic, which will accelerate the process of historical pollutants entering the global cycle and have a certain impact on the global ecosystem.

Therefore, we need to control the pollution of important emission sources of dioxins and seek possible methods to degrade them into low-toxicity or nontoxic products.

Per- and poly fluoroalkyl substances (PFASs)

PFASs have been comprehensively investigated in various environmental and biological matrices in the Arctic, while very limited studies and datasets are available for the Antarctic. Generally, neutral PFASs mostly partition to air and undertake global transport via the atmosphere and are present in the Arctic and Antarctic. The degradation of neutral PFASs in air and biota have become a major source for the presence of perfluoroalkylated sulfonate (PFSA) and carboxylic acid (PFCA) in the polar regions. Data on neutral PFASs in seawater and biota are rare. Ionic PFASs were mainly transported via ocean currents, and PFASs in the Arctic were one order of magnitude higher than those in the Southern Ocean. Perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) were the predominant PFASs in seawater and snow, while perfluoroundecanoic acid (PFUnDA), perfluorononanoic acid (PFNA), PFOS and perfluorobutane sulfonate (PFBS) were the major congeners in biota.

Reemission of neutral PFASs from snow to air has been observed in the Arctic. Glaciers, sea ice and snow can release PFASs accumulated through the winter or past years into coastal waters of the polar region. Several novel PFASs have been found in the environment and biota in the past ten years, such as hexafluoropropylene oxide dimer acid (HFPO-DA) in seawater and snow, 6:2, 8:2 fluorotelomersulfonate (6:2 and 8:2 FTS) in runoff near airports, 6:2 chlorinated polyfluorinated ether sulfonate (6:2 Cl-PFESA) in lake water and polar bears, and fluorotelomer phosphate diesters (6:2, 8:2 DiPAP) in bird eggs.

Considering the large number of PFASs on the global market, it could be expected that more alternative PFASs will be identified from extractable organic fluorinated substances and discovered in environmental matrices. Therefore, further study need to be conducted to elucidate the spatial and temporal trends of the novel PFASs in polar regions. Given the concern of climate change and enhanced human activities in the polar regions, long-term monitoring programs and modeling exercises should be considered to improve the knowledge for understanding the environmental fate and toxicity and regulatory plans for the management of PFASs.

Organophosphate esters (OPEs)

OPEs have been investigated in various environmental matrices, such as air, water, sediment and organisms, in the Arctic and Antarctic. Available data in the Arctic indicated OPEs are the predominant chemical group in both the environment and the biota. Long-range environmental transport (LRET) with atmosphere and ocean currents are the major pathways in which OPEs are transported to polar regions. However, enhanced human activities, such as research stations, tourism and shipping, can directly contribute to the local environment. Obviously, this is attributed to the high concentrations of OPEs measured in the Antarctic. Therefore, background measurements of OPEs need to be considered to minimize the interferences. The occurrence of OPEs in Svalbard, such as in the Arctic fox and soil, highlights that comprehensive surveys of OPEs in terrestrial environments need to be included in long-term monitoring programs. In addition, the degradation of OPEs under nutrient-limited conditions suggests that these can be a source of new P to the polar regions, which requires comprehensive characterization.

Apart from novel OPEs, some metabolites of OPEs might be more toxic than their parent compounds (Fu et al., 2021). Therefore, future studies should assess the occurrence, environmental behavior, and ecological risk of OPEs and their metabolites in polar regions. Overall, given the pressure of increasing human activities and climate change, future research

needs to couple the observations and models to elucidate the various biogeochemical and geophysical processes of OPEs in the polar environment.

Pharmaceuticals and personal care products (PPCPs)

The literature has shown the wide presence of PPCPs in both Arctic and Antarctic environments. High concentrations of pharmaceuticals are usually found in coastal areas receiving effluents from wastewater treatment plants (WWTPs) at research bases and local settlements. Therefore, the concentrations of PPCPs in water, sediment and organisms should be investigated. The data on PPCPs in organisms are quite limited. More studies on the bioaccumulation and toxicities of PPCPs to marine organisms need to be conducted in future research programs. Apart from local emissions, personal care products (PCPs), such as synthetic fragrance materials (FMs), ultraviolet light filter agents (UV-Fs) and cyclic volatile methylsiloxanes (cVMS), can be transported with air from low and middle latitudes to polar regions. It is necessary to include volatile and semi-volatile PCPs in existing atmospheric monitoring programs.

PPCPs are mostly stable and can be released directly in sewer systems, however, they may also be degraded into transformation products (Kallenborn et al., 2018). Consequently, both the target parent compounds and the major transformation products must be included in future monitoring programs and considered for comprehensive risk assessment (Daughton, 2003). There is still a large knowledge gap in research regarding the long-term effects and cocktail effect of PPCPs in polar regions. The ultra-trace concentration levels of PPCPs in seawater, sediments and organisms in polar environments is a significant challenge for existing analytical methods. However, technological developments and advances in new applications will further reduce quantification constraints and lead to the identification of new and currently unidentified PPCPs (Kallenborn et al., 2018).

Presently, international or national regulations are absent to control the application of PPCPs in polar regions. There is still a large knowledge gap in research regarding the long-term effects and cocktail effect of PPCPs. Relevant PPCPs should be implemented into already established long-term international, national and regional monitoring programs to serve as a scientific basis for region-specific "watch lists" for PPCPs, particularly in transition coastal areas of the Arctic and Antarctic.

Zusammenfassung

Klassische (persistente) und neu auftretende organische Schadstoffe (POPs und EOCs) in Polarregionen sind aufgrund ihrer Persistenz, Bioakkumulation und ihres Toxizitätspotenzials zu erheblichen Bedenken geworden. Der Klimawandel kann den biogeochemischen Kreislauf von POPs und EOCs verändern und ihre Auswirkungen auf polare Ökosysteme verstärken. Die globale Industrialisierung und der Wiederaufbau von Forschungsstationen betreffen sowohl die Arktis als auch die Antarktis, was zu einer Zunahme menschlicher Aktivitäten wie Tourismus, Schifffahrt, Ressourcenerkundung und dem Bau von Forschungsstationen geführt hat. Das Auftreten von POPs und EOCs durch Ferntransport und lokale Abladungen hat Auswirkungen auf empfindliche polare Ökosysteme hinterlassen. Daher sind dringend Maßnahmen erforderlich, um zeitliche Trends von POPs zu beobachten und neue EOCs in Polarregionen zu untersuchen.

Das Arctic Monitoring and Assessment Program (AMAP) wurde 1991 gegründet, um Komponenten der Arctic Environmental Protection Strategy (AEPS) umzusetzen und ist zu einer Programmgruppe des Arktischen Rates geworden. Das AMAP hat mehrere Forschungsprogramme zur Überwachung klassischer POPs und EOCs in Luft, Sedimenten und Organismen in der gesamten Arktis gestartet. In den letzten Jahrzehnten hat das AMAP regelmäßige Bewertungen für klassische Schadstoffe und neu auftretende Chemikalien veröffentlicht, die von Bedenken für Arktis und Klimawandel sein könnten. Wissenschaftliche Informationen wurden übermittelt, um Interessenvertreter der arktischen Regierungen bei der Ergreifung von Hilfs- und Vorbeugungsmaßnahmen bezüglich Schadstoffen zu unterstützen. Für die Antarktis hat der Wissenschaftliche Ausschuss für Antarktisforschung (SCAR) eine lange und erfolgreiche Geschichte bei der Zusammenfassung von politisch relevanten wissenschaftlichen Erkenntnissen für politische Entscheidungsträger durch seine Arbeitsgruppen, wie die Input Pathways of Persistent Organic Pollutions to Antarctic (ImPACT). Die Erfahrungen des AMAP wurden bei Forschungsprojekten und Aktivitäten von ImPACT erfolgreich für die Beobachtung und Bewertung chemischer Schadstoffe in der Antarktis transferiert.

Die Daten zu konventionellen POPs in Umweltmedien und Biota zeigen aufgrund weltweiter Bemühungen, ihre Herstellung und Verwendung zu verbieten, sowohl in der Arktis als auch in der Antarktis rückläufige Trends. Es wurde jedoch eine Reemission von POPs beobachtet, die sich zuvor in der polaren Umgebung angesammelt haben, und diese POPs können nach den durch die globale Erwärmung verursachten Prozessen des Eisrückgangs, des Gletscherschmelzens und des Permafrostauftauens wieder in den globalen Kreislauf eintreten. Daher sollte in Polargebieten eine kontinuierliche Beobachtung von konventionellen POPs durchgeführt werden. Es wurden jedoch eine Reihe von EOCs in verschiedenen Umweltmatrizes in der Arktis und Antarktis gemeldet. Einige EOCs, wie Per- und polyfluorierte Alkylverbindungen (PFASs), Chlorparaffine (SCCPs und MCCPs) und Organophosphatester (OPEs), wurden in langfristige Atmosphärenüberwachungsprogramme des AMAP aufgenommen. Im Rahmen nationaler und regionaler Forschungsprogramme wurden Screening-Untersuchungen zu EOC in der Umwelt und in biologischen Matrizes durchgeführt. Daten über EOC in der Antarktis sind selten. Dennoch wurde der weiträumige Transport von EOC in der Umwelt durch ihr Vorkommen in Eiskernen, Schnee und Seewasser in den Polarregionen aufgezeigt. Daher muss die Untersuchung von EOCs in der Antarktis durch nationale und internationale Forschungsprogramme verstärkt werden.

Umweltprobenbanken (ESBs) für polare Arten wurden mit Unterstützung nationaler Forschungsprogramme eingerichtet, insbesondere in den arktischen Staaten. Diese Banken bieten die beste Chance, den historischen Fingerabdruck chemischer Schadstoffe zu rekonstruieren, die in der polaren Umgebung aufgezeichnet wurden. Allerdings ist der ESB für die Antarktis recht begrenzt. Daher sollte ein langfristiges Überwachungsprogramm in der Antarktis initiiert werden, um typische Umwelt- und biologische Proben zu sammeln, um einen antarktischen ESB zu etablieren.

Eine Bewertung der Wechselwirkung zwischen dem arktischen Klimawandel, POPs und Chemikalien von zunehmender Bedeutung wurde von der AMAP von 2019 bis 2021 unter einzigartigen Veränderungen der arktischen Schnee-, Wasser-, Eis- und Permafrostbedingungen durchgeführt. Konventionelle POPs wie Organochlorpestizide (OCPs) und polychlorierte Biphenyle (PCBs) zeigten eine starke Reaktion auf bestimmte Klimawandelszenarien. Gletschereis und -schnee fungierten als sekundäre Emissionsquellen in den Polarregionen und setzten POPs und EOCs in Atmosphäre und Ozean frei. Daher muss die zukünftige Forschung die verschiedenen biogeochemischen und geophysikalischen Prozesse unter Klimawandel und anthropogenen Belastungen verstehen, um das Umweltverhalten und das Toxizitätsrisiko von EOCs in Polarregionen vorhersagen zu können.

Polybromierte Diphenylether (PBDEs)

Obwohl die Kontrollen der Anwendung von Penta- und Octa-PBDE-Produkten allmählich zu einem Rückgang der Konzentrationen in Umweltproben führen, ist es nach wie vor dringend erforderlich, eine kontinuierliche Erhebung der zeitlichen Trends von PBDE in der polaren Umwelt durchzuführen, wenn der Status der PBDE-Verschmutzung ungewiss ist. Es werden daher mehr Daten über die zeitliche Variabilität der PBDE-Konzentrationen und -Verteilungen in Umweltmatrizen und an Probenahmestellen benötigt. PBDE werden an Land durch menschliche Aktivitäten erzeugt, mit sekundären Emissionen aus kontaminierten Wasserreservoirs. Es gibt jedoch kaum Berichte über den PBDE-Austausch zwischen Luft und Oberfläche in der Arktis/Antarktis, was unser Verständnis darüber erschwert, ob frühere "Senken" zu Quellen werden. Im Zusammenhang mit dem Klimawandel sind die Auswirkungen der globalen Erwärmung auf den Transport und das Verhalten von PBDE in den Polargebieten sowie die zeitliche Tendenz und die Bioakkumulation in Biota weitgehend unerforscht. In Anbetracht einiger Hinweise auf eine Anreicherung von Decabromdiphenylether (BDE-209) in antarktischen Biota verdient der weiträumige Transport und die Ablagerung von BDE-209 in der Antarktis mehr Aufmerksamkeit. Da neuartige bromierte Flammschutzmittel als PBDE-Ersatz vermarktet werden, wird eine Untersuchung des Vorkommens und der Emissionstrends dieser aufkommenden Alternativen in der polaren Umwelt empfohlen.

Polycyclische aromatische Kohlenwasserstoffe (PAKs)

Es fehlt nach wie vor eine breit angelegte räumliche Analyse der PAKs und insbesondere ihrer Derivate aus der terrestrischen und marinen Umwelt in den Polarregionen. Neuere Studien zu den zeitlichen Trends von PAK aus der Luftüberwachung und aus datierten Bohrkernen sind begrenzt. Polare PAKs, wie sauerstoffhaltige und nitrierte Verbindungen haben eine stärkere genotoxische, mutagene und karzinogene Wirkung als ihre Ausgangs-PAK. Die Auswirkungen auf die Umwelt und die menschliche Gesundheit sowie die Bewertung der relativen primären und sekundären Eingänge von PAK-Derivaten in den Polarregionen sind jedoch noch weitgehend unerforscht. Mehrere PAK werden nach dem photochemischen Prozess in langlebige Derivate umgewandelt und können einen weiträumigen atmosphärischen Transport (LRAT) durchlaufen. Die Bewertung des LRAT-Potenzials von PAK-Derivaten, insbesondere von Nitro-PAKs, sollte erweitert werden. Über die Beeinflussung atmosphärisch transportierter PCBs durch biologische Pumpen in der Arktis wurde berichtet. Vor dem Hintergrund langfristig möglicherweise abnehmender PAK-Emissionen aus primären Quellen sowie der globalen Erwärmung könnte der biologische Pumpeffekt jedoch durch die zunehmende Freisetzung von PAK in die Luft aus lokalen sekundären Quellen und die Migration von lange konservierten PAK aus dem Meereis in das Wasser als Folge der verstärkten Eisschmelze abgeschwächt werden. Die Verbindung zwischen dem Umweltverhalten von PAK in den Polarregionen und dem Klimawandel oder anthropogenen Aktivitäten verdient langfristige Aufmerksamkeit.

Organochlorpestizide (OCPs)

Die gewebespezifischen Protein- und Lipidgehalte spielen zusammen mit zahlreichen anderen biochemischen Prozessen eine wichtige Rolle bei der Toxikokinetik und dem Verbleib von chlorierten und bromierten Kontaminanten, Stoffwechsel- und Naturprodukten. Eine verstärkte, selektive Retention bestimmter Organohalogenklassen in bestimmten Geweben/Körperkompartimenten kann daher zu standortspezifischen toxikologischen Wirkungen und unerwünschten Wirkungen bei hochkontaminierten Svalbard-Eismöwen führen. Weitere Studien sind gerechtfertigt, um die Mechanismen der Bindungswechselwirkungen von Makromolekülen und die Akkumulation von Organohalogenen in Vogelarten zu untersuchen (Verreault et al. 2007). Seitdem in den 1970er und 1980er Jahren damit begonnen wurde, ältere Pestizide wie Dichlordiphenyltrichlorethan (DDT) zu regulieren, hat der Einsatz von Ersatzchemikalien, die ähnlichen Zwecken dienen (d.h. Verringerung der Exposition gegenüber durch Insekten übertragenen Krankheiten, Schutz von Ernten und Aufrechterhaltung der landwirtschaftlichen Erträge, massiv zugenommen). Theoretisch können derzeit verwendete Pestizide (CUPs) zur Verwendung zugelassen werden, wenn keine Persistenz in der Umwelt lange nach der Verwendungsdauer beobachtet wird und wenn die Chemikalien ein geringes Bioakkumulationspotenzial haben; Daher sollten CUPs weniger Umweltauswirkungen haben als die alten Pestizide, die sie ersetzen. Trotz ihrer geringeren Konzentrationen in Biota der Arktis im Vergleich zu älteren OCPs wurden Biomodifikationseffekte gefunden, und ihr LRT-Potenzial wurde ebenfalls nachgewiesen; Daher ist die kontinuierliche Überwachung sowohl bekannter als auch potenzieller arktischer Schadstoffe wichtig, um sicherzustellen, dass CUPs und ihre Ersatzchemikalien minimale Auswirkungen auf die Gesundheit von Mensch und Umwelt haben.

Polychlorierten Biphenylen (PCB)

Deutlich wird, dass PCBs in Umweltmedien und Biota sowohl in der Arktis als auch in der Antarktis aufgrund der weltweiten Bestrebungen, die Herstellung und Verwendung von PCB zu verbieten, eine rückläufige Tendenz aufweisen. Allerdings dürften durch den Klimawandel zuvor in der Umwelt abgelagerte PCB wieder in den globalen Kreislauf gelangen. Daher sollten wir dem Verhalten von PCB in Polargebieten weiterhin große Aufmerksamkeit schenken. Wir haben eine Reihe bestehender Studien überprüft und mehrere Wissenslücken in der wissenschaftlichen Forschung, Entscheidungsfindung und Managementpolitik zusammengefasst. Die Kontamination der Arktis absichtlich hergestellte PCBs ist gut dokumentiert und die jüngste Entdeckung von unbeabsichtigt hergestellten (UP) PCBs wie PCB-5 und PCB-11 haben zunehmend Anlass zur Sorge gegeben (Bartlett et al., 2019). PCB-11 wurden im Oberflächenschnee von Lomonosovfonna, Svalbard, in den Jahren 2009–2010 und 2013–2014 bestimmt und machten 4% der gesamten PCB aus (Garmash et al., 2013). PCB-11 und andere UP-PCBs werden in der Arktis nicht gut verstanden. Obwohl ihre Konzentrationen nicht so hoch waren wie die einiger alter PCBs, ist die Toxizität der Ausgangsquelle und der Metaboliten noch unklar, was zu einem höheren negativen Risikoeffekt führen könnte.

In den meisten früheren Forschungen wurden etwa 2 oder 3 Dekaden von PCB-Kongeneren untersucht, die <10% der gesamten PCB ausmachen. Andere UP-PCBs in Polargebieten sind weniger bekannt, wie z. B. PCB-11, das in neueren Studien zu 8,5% der gesamten PCBs beigetragen hat (Bartlett et al. 2019); Daher sind weitere Messungen aller 209 PCB-Kongenere unerlässlich, wenn wir beginnen wollen, ihre Beteiligung an der Umweltverschmutzung zu

verstehen. Darüber hinaus empfehlen wir für die zukünftige Risikoüberwachung, die synergistischen toxischen Wirkungen mehrerer Schadstoffe zu untersuchen und die Lücken in der räumlichen Verteilung von Daten zu schließen.

Die Transportwege von POPs in die Arktis und Antarktis sind nicht ausreichend verständlich und leiden unter einem Mangel an räumlichen und zeitlichen Beobachtungen. Viele Modelle wurden entwickelt, um das Verhalten und den Transport von PCB aus den Quellregionen in die Polargebiete vorherzusagen. Die Genauigkeit dieser Modelle hängt jedoch von der Auflösung der geografischen Karte, der Dichte meteorologischer Beobachtungen und den physikalischchemischen Prozessen (Wolken, Nebel, Niederschlag, Temperatur und Sonnenlicht sowie Luft-Meer-Austausch und Gas-Partikel-Austausch usw.), deren Einflüsse in den Modellen unzureichend erfasst werden. In Bezug auf die terrestrischen Pfade und Süßwasserpfade von POPs in die Arktis ist die Forschung zum Luft-Schneedecken-Austausch, zum Fluss des Inputs von Festland und Süßwasser in den Ozean, zur Sedimentresuspension und zum Einfluss der Eisbedeckung kaum bekannt. Darüber hinaus haben Modelle der Ozean- und Eiszirkulation derzeit oft eine zu geringe Auflösung, um Grenzströmungen und Wirbel zu erfassen, die beide wahrscheinlich die Hauptwege des Schadstofftransports sind. Darüber hinaus treten Schadstoffe nur als passive Tracer in die Modelle ein, und zukünftige Modellierungsbemühungen müssen realistischere Schadstoffeigenschaften berücksichtigen, einschließlich Aufnahme durch Biota, Partikelabsorption und kinetische Verluste.

Die Festlegung von Konzentrationsgrenzwerten ist sowohl für einzelne Verbindungen als auch für realistische Zusammensetzungen erforderlich, die zwar auf eine biologische Relevanz hindeuten, aber noch nicht statistisch erfasst wurden. Einerseits besteht ein anhaltender Bedarf, Konzentrationsschwellen für biologisch relevante Gesundheitseffekte bei Wildtieren und Fischen in Polargebieten festzulegen und zu verfeinern. Andererseits besteht Bedarf an einer besseren Identifizierung der kumulativen und interaktiven Wirkungsschwellen der Schadstoffexposition. Die multiplen Stressoren vieler POPs und des Klimawandels sollten untersucht werden, um die tatsächlichen Gesundheitsrisiken für Mensch und Tier abzuschätzen (Dietz et al. 2019; Letcher et al. 2010; Szopińska et al. 2019).

Chlorparaffine (CP)

Derzeit ist die Forschung zum Umweltverhalten von kurzkettige Chlorparaffine (SCCPs) weitgehend durch ihre quantitative Analyse begrenzt, und die bestehenden Analysemethoden können SCCP-Monomere nicht einzeln untersuchen und analysieren. Bei der Auswahl von Standardprodukten nehmen die meisten Labore hauptsächlich den kommerziellen industriellen Produktmix als Standard. Die meisten SCCPs, die in die Umwelt freigesetzt werden, unterliegen jedoch einer selektiven Umweltmigration in die Umwelt und einer biologischen Stoffwechselumwandlung, und ihre Zusammensetzung kann sich von der kommerzieller Mischungen erheblich unterscheiden. Dieser Unterschied spiegelt sich schließlich in der Inkonsistenz der Eigenschaften von SCCPs zwischen dem analytischen Standard und der tatsächlichen Probe wider, was dann auf die Differenz der Reaktion des Gerätedetektors auf verschiedene SCCPs übertragen wird, was zu Analysefehlern führt. Aufgrund der Unterschiede in den Proben, die von verschiedenen Labors, Analysestandards, Vorbehandlungs- und instrumentellen Analysemethoden im Analyseprozess bestimmt wurden, ist es schwierig, SCCP horizontal zwischen Studien zum Vorkommen in Ökosystemen und zum Umweltverhalten zu vergleichen. Daher ist es dringend erforderlich, geeignete Analysemethoden mit höherer Sensitivität und Genauigkeit zu entwickeln.

Polychlorierte Naphthaline (PCNs)

Obwohl das Vorkommen von PCNs in einer Reihe von Medien nachgewiesen wurde, darunter Luft, Biota und Säugetiere, fehlte es an systematischen Überwachungsprojekten, um die zeitliche Diversifizierung von PCNs zu untersuchen und die Auswirkungen der Umsetzung der Stockholmer Konvention zu bewerten. Nach unserem Wissen umfassen die Quellen von PCN hauptsächlich atmosphärische Wanderungen über große Entfernungen und anthropogene Emissionsquellen, aber der Anteil jedes Typs ist nicht klar. Daher sollte für die spezifische Probenahmestelle zur Untersuchung ein wenig kontaminierter Standort ausgewählt werden, und sogar die von der Messstation selbst erzeugten anthropogenen Emissionen sollten berücksichtigt werden.

Darüber hinaus kennen wir die toxischen Veränderungen von PCN-Abbauprodukten in speziellen polaren Umgebungen nicht, sodass es notwendig ist, die Abbauprodukte von PCN zu analysieren, zu überwachen und umfassend zu charakterisieren. Frühere Studien untersuchten wenige polare biologische Arten und es fehlte an systematischer umfassender Forschung zu verschiedenen Arten in der Struktur der Nahrungskette, sodass der Biomagnifikationseffekt von PCNs in der polaren Nahrungskette weiterer Untersuchungen wert ist. Im heutigen globalen Erwärmungsszenario führt das Schmelzen von Eis und Schnee zur schrittweisen Freisetzung gespeicherter POPs. Daher ist es notwendig, die biogeochemischen und geophysikalischen Prozesse dieser POPs (einschließlich PCNs) kontinuierlich zu beobachten, um ihren Verbleib in der Umwelt und ihre toxikologischen Risiken vorherzusagen.

Dioxine

Der Nachweis in diesen abgelegenen Gebieten zeigt, dass sie sich in Polargebieten kontinuierlich anreichern können, insbesondere für hochchlorierte Chlorierte Dibenzodioxine und Dibenzofiurane (PCDDs/PCDFs), die sich in diesen Gebieten stabil ansammeln können. Daher ist es notwendig, den zeitlichen Änderungstrend dieser Schadstoffe zu beachten. Durch systematische Forschung und Überwachung können wir diesen Schadstoffen kontinuierlich Aufmerksamkeit schenken und ein Modell zur Vorhersage des zukünftigen Umweltverhaltens erstellen. Als Nebenprodukt unvollständiger Verbrennungsreaktionen und der chemischen Produktion werden Dioxine immer noch kontinuierlich produziert, aber das Ausmaß der Nutzung von Verbrennungsreaktionen nimmt schnell zu; Darüber hinaus steigt die Nachfrage nach chemischen Produkten rapide an, sodass die Entstehung von Dioxinen nicht vermieden werden kann. Die Erderwärmung führt in der Antarktis und der Arktis kontinuierlich zum Abschmelzen von Eis und Schnee, was den Eintritt historischer Schadstoffe in den globalen Kreislauf beschleunigt und dies wird gewisse Auswirkungen auf das globale Ökosystem haben. Daher müssen wir die Belastung von wichtigen Emissionsquellen von Dioxinen kontrollieren und nach Methoden suchen, um sie zu Produkten mit geringer Toxizität oder ungiftigen Produkten abzubauen.

Per- und polyfluorierte Alkylsubstanzen (PFAS)

PFAS wurden umfassend in verschiedenen Umwelt- und biologischen Matrizes in der Arktis untersucht, während für die Antarktis nur sehr begrenzte Studien und Datensätze verfügbar sind. Im Allgemeinen verteilen sich neutrale PFAS hauptsächlich in der Luft, machen einen globalen Transport über die Atmosphäre durch und sind in der Arktis und Antarktis vorhanden. Der Abbau von neutralen PFAS in Luft und Biota ist zu einer Hauptquelle für das Vorhandensein von perfluorierten Alkylcarbonsäuren (PFCA) und perfluorierten Alkylsulfonsäuren (PFSA) in den Polarregionen geworden. Daten zu neutralen PFAS in Meerwasser und Biota sind rar. Ionische PFAS wurden hauptsächlich über Meeresströmungen transportiert, und die PFAS in der Arktis waren um einiges höher als im Südpolarmeer. Perfluoroctansulfonsäure (PFOS) und Perfluoroctansäure (PFOA) waren die vorherrschenden PFAS in Meerwasser und Schnee, während Perfluorundecansäure (PFUnDA), Perfluornonansäure (PFNA), PFOS und Perfluorbutansulfonsäure (PFBS) die wichtigsten Kongenere in Biota waren.

Die Reemission von neutralen PFAS aus Schnee in die Luft wurde in der Arktis beobachtet. Gletscher, Meereis und Schnee können im Winter oder in den vergangenen Jahren angesammelte PFAS in die Küstengewässer der Polarregion freisetzen. In den letzten zehn Jahren wurden mehrere neuartige PFAS in der Umwelt und in Biota gefunden, wie Ammonium Perfluor-2-Propoxypropansäure (HFPO-DA) in Meerwasser und Schnee, Fluortelomersulfonate (6:2, 8:2 FTS) im Abfluss in der Nähe von Flughäfen, chloriertes polyfluoriertes Ethersulfonat (6:2 Cl-PFESA) in Seewasser und Eisbären und Fluortelomerphosphatdiester (6:2, 8:2 DiPAP) in Vogeleiern.

Angesichts der großen Anzahl von PFAS auf dem Weltmarkt ist zu erwarten, dass weitere alternative PFAS aus extrahierbaren organischen fluorierten Substanzen identifiziert und in Umweltmatrizes entdeckt werden. Daher müssen weitere Studien durchgeführt werden, um die räumlichen und zeitlichen Trends der neuartigen PFAS in Polarregionen aufzuklären. Angesichts der Besorgnis über den Klimawandel und verstärkte menschliche Aktivitäten in den Polarregionen sollten langfristige Überwachungsprogramme und Modellierungsübungen in Betracht gezogen werden, um das Wissen zum Verständnis des Verbleibs und der Toxizität in der Umwelt und der Regulierungspläne für den Umgang mit PFAS zu verbessern.

Organophosphatester (OPEs)

OPEs wurden in verschiedenen Umweltmatrizes wie Luft, Wasser, Sediment und Organismen in der Arktis und Antarktis untersucht. Verfügbare Daten in der Arktis weisen darauf hin, dass OPEs die vorherrschende chemische Gruppe sowohl in der Umwelt als auch in der Biota sind. Langstrecken-Umwelttransport (LRET) mit Atmosphäre und Meeresströmungen sind die Hauptwege, auf denen OPEs in Polarregionen transportiert werden. Zunehmende menschliche Aktivitäten wie Forschungsstationen, Tourismus und Schifffahrt können jedoch direkt zur lokalen Umwelt beitragen. Dies führt offenbar zu hohen Konzentrationen von OPEs in der Antarktis. Daher müssen Hintergrundmessungen von OPEs in Betracht gezogen werden, um die Interferenzen zu minimieren. Das Vorkommen von OPEs in Svalbard, wie z. B. im Polarfuchs und im Boden, macht deutlich, dass umfassende Untersuchungen von OPEs in terrestrischen Umgebungen in langfristige Überwachungsprogramme aufgenommen werden müssen. Darüber hinaus deutet der Abbau von OPEs unter nährstoffbegrenzten Bedingungen darauf hin, dass diese eine Quelle für neuen Phosphor in den Polarregionen sein können, was eine umfassende Charakterisierung erfordert.

Abgesehen von neuartigen OPEs können einige Metaboliten von OPEs toxischer sein als ihre Ausgangsverbindung (Fu et al. 2021). Daher sollten zukünftige Studien das Vorkommen, das Umweltverhalten und das ökologische Risiko von OPEs und ihren Metaboliten in Polarregionen bewerten. Insgesamt muss angesichts des Drucks zunehmender menschlicher Aktivitäten und des Klimawandels die zukünftige Forschung die Beobachtungen und Modelle koppeln, um die verschiedenen biogeochemischen und geophysikalischen Prozesse von OPEs in der polaren Umgebung aufzuklären.

Pharmazeutika und Körperpflegeprodukte (PPCPs)

Die Literatur hat die breite Präsenz von PPCPs sowohl in arktischen als auch in antarktischen Umgebungen gezeigt. Hohe Konzentrationen von Arzneimitteln werden normalerweise in Küstengebieten gefunden, die Abwässer von Kläranlagen in Forschungsbasen und lokalen Siedlungen erhalten. Daher sollten die Konzentrationen von PPCPs in Wasser, Sediment und Organismen untersucht werden. Die Daten zu PPCPs in Organismen sind recht begrenzt. Weitere Studien zur Bioakkumulation und Toxizität von PPCPs für Meeresorganismen müssen in zukünftigen Forschungsprogrammen durchgeführt werden. Abgesehen von lokalen Emissionen können Körperpflegeprodukte (PCPs) wie synthetische Duftstoffe (FMs), UV-Lichtfilter (UV-Fs) und zyklische flüchtige Methylsiloxane (cVMS) mit der Luft aus niedrigen und mittleren Breiten in Polarregionen transportiert werden. Es ist notwendig, flüchtige und halbflüchtige PCPs in bestehende Atmosphärenüberwachungsprogramme aufzunehmen.

PPCPs sind meist stabil und können direkt in Kanalisationen freigesetzt werden; sie können aber auch zu Transformationsprodukten abgebaut werden (Kallenborn et al. 2018). Folglich müssen sowohl die Zielausgangsverbindungen als auch die wichtigsten Transformationsprodukte in zukünftige Überwachungsprogramme aufgenommen und für eine umfassende Risikobewertung berücksichtigt werden (Daughton 2003). Es besteht noch eine große Wissenslücke in der Forschung zu den Langzeitwirkungen und Cocktaileffekten von PPCPs in Polarregionen. Die Ultraspurenkonzentration von PPCPs in Meerwasser, Sedimenten und Organismen in polaren Umgebungen ist eine erhebliche Herausforderung für bestehende Analysemethoden. Fortschritte in der technologischen Entwicklung und neue Applikationen werden die Quantifizierungsgrenzen jedoch weiter verringern und zusätzlich zur Identifizierung neuer und derzeit nicht identifizierter PPCP führen (Kallenborn et al. 2018).

Gegenwärtig fehlen internationale oder nationale Vorschriften, um die Anwendung von PPCPs in Polarregionen zu kontrollieren. Es besteht noch eine große Wissenslücke in der Forschung bezüglich der Langzeitwirkung und Cocktailwirkung von PPCPs. Relevante PPCP sollten in bereits etablierte langfristige internationale, nationale und regionale Überwachungsprogramme implementiert werden, um als wissenschaftliche Grundlage für regionenspezifische "Beobachtungslisten" für PPCP zu dienen, insbesondere in Übergangsküstengebieten der Arktis und Antarktis.

1 Introduction

Pristine polar ecosystems have been impacted by numerous natural and anthropogenic factors. Increasing human activities and a changing climate pose major pressures on the polar environment (AMAP 2017; 2021). The direct and indirect inputs of man-made pollutants from local emissions and remote continental sources have led both the Arctic and the Antarctic to act as ultimate sinks, which can cause unexpected disturbances and risks to polar ecosystems and create a global problem for the future Earth.

With the rapid development of economy, industry and population in the world, many chemical pollutants are introduced into the environment in large quantities (Brown and Wania 2008; Cabrerizo et al., 2013; Wania 2007). POPs are typical synthetic chemicals and ubiquitous in the global environment. They have common characteristics of persistence, toxicity, bioaccumulation potential and tendency of long-range environmental transport (LRET) (Arnot et al., 2011; Nash et al., 2017). Therefore, POPs are considered to pose a threat to environmental and human health and are subject to the Stockholm Convention's requirements to reduce and ultimately eliminate these compounds from the environment. Initially, twelve POPs were recognized as causing adverse effects on humans and the ecosystem, including organic pesticides (aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, toxaphene), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDDs/PCDFs). Subsequently, sixteen new chemicals were added to the Stockholm Convention until 2019 (SC, 2022). More organic chemicals are under evaluation and will be reviewed by the POP Review Committee.

In addition to the classic POPs, several organic chemicals have been found in the polar regions as emerging organic contaminants (EOCs). Generally, emerging contaminants are expected to be chemicals that show the potential to pose risks to human health or the environment but are not yet subjected to regulatory criteria for the protection of human health or the environment (Sauve and Desrosiers 2014). EOCs include alternative brominated flame-retardants (BFRs), short-chain PFASs and their precursors, pharmaceuticals and personal care products (PPCPs), organophosphate esters (OPEs), phthalate esters (PAEs), chlorinated paraffins (CPs) and cyclic volatile methylsiloxanes (cVMS). Various adverse effects caused by EOCs in the environment have been reported in the literature.

Volatile and semi-volatile organic contaminants undergo LRET with air and/or particles. However, polar organic contaminants may tend to undergo LRET with oceanic circulations and marine particulates (Nash 2011). These chemicals can reach the polar regions through global distillation and fractionation processes (Wania 2007; Wania and Dugani 2003). Therefore, both the Arctic and the Antarctic have accumulated large quantities of organic contaminants and have become important sinks for global POPs. Consequently, the presence of novel organic contaminants in polar environments and organisms can provide direct evidence for their potential for LRET and bioaccumulation. Furthermore, human activities in polar regions have changed and are rapidly increasing, e.g., intensive expeditions, blooming of polar tourisms and shipping. The direct release of organic contaminants from local research stations was also observed in both the Arctic and the Antarctic. 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).

The working group of Input Pathways of persistent organic pollutants to Antarctic (ImPACT) under the Scientific Committee on Antarctic Research (SCAR) was proposed to facilitate the coordinated investigation and monitoring of chemical input to the Antarctic region. The research activities and findings for POPs in the Antarctic have been reviewed (Nash 2011). Similarly, the

Arctic Environmental Protection Strategy (AEPS) was signed by eight Arctic countries in 1991, which proposed monitoring the levels and assessing the effects of anthropogenic pollutants in all compartments of the Arctic, as organized by the Arctic Monitoring and Assessment Programme (AMAP). To date, the AMAP has published several assessment reports for POPs and emerging organic contaminants (EOCs) in the Arctic (AMAP 2004; 2017; 2021). Terrestrial animals, marine mammals and circumpolar Inuit in the Arctic are highly affected by organic contaminants because of geographic locations, lipid-rich diets, and their placement at the top of the food chain (Nash 2011). The organic contaminants found in the Antarctic mainly represent the emissions from the Southern Hemisphere, which has a relatively low population and historical industrial level. In both Arctic and Antarctic, many research stations have been found up for meteorology and environmental monitoring, especially for atmospheric pollutions. These facilities provide unique support for investigation of POPs nad EOCs in the pan Arctic and Antarctic (Figure 1).

In this report, we review the levels of both classic POPs and EOCs in environmental and biological matrices in polar regions; these include PFASs, brominated flame retardants (BFRs), organophosphate esters (OPEs), OCPs, PCBs, PAHs, chlorinated paraffins (CPs), polychlorinated naphthalene (PCN) and PPCPs. In addition, studies on atmospheric deposition, air-water exchange and remobilization of EOCs were summarized for a better understanding of the environmental pathways of EOCs in polar regions. Consequently, research gaps and future perspectives were identified for important research and monitoring efforts required for assessing the impact of organic contaminants in the changing polar regions.



Figure 1: Research stations for environmental monitoring in the Arctic and Antarctic

Source: Hereon.

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2 Polybrominated diphenyl ethers (PBDEs)

2.1 Introduction

Polybrominated diphenyl ethers (PBDEs) have been used extensively as flame-retardants in a variety of consumer products from the 1970s to the early 2000s. Their occurrence has been confirmed in various environmental matrices, even in polar regions. Aroused concerns of PBDE persistence, possible adverse effects on wildlife and humans, and bioaccumulation potential lead to the increasing amount of attention focused on PBDEs.

From 1970 to 2005, the total historical production of all PBDEs was estimated to range from 1.3 million to 1.5 million tons, consisting of ca. 100,000 tons of penta-BDEs, 100,000 tons of octa-BDEs, and over 1.1 million tons of deca-BDEs (UNEP, 2010a). Based on BSEF (2006), the rapid global demand for PBDEs since the 1970s triggered a PBDE global production amount of ca. 40 kt in 1992, and this value increased to approximately 67 kt in 2001(Earnshaw et al., 2013). Three main technical PBDE mixtures have been produced and marketed at various degrees of bromination and include deca-BDE, octa-BDE, and penta-BDE (Darnerud et al., 2001). The Asian shares of the global market are 0%, 52%, and 42% for penta-, octa- and deca-BDE products, respectively (de Wit, 2002), while PBDE productions in Europe were expected to have ceased in the late 1990s, with the quantity of PBDE produced and imported as chemical mixtures totaling ca. 7 to 11 kt from 1986 to 1989. The production of penta- and octa-BDEs was voluntarily discontinued in the U.S. until 2005; however, concrete data on PBDE production in the U.S. are not publicly accessible (Choy et al., 2010; de Wit et al., 2010). Regarding PBDE application, more than 95% of total penta-BDE consumption occurs in North America, and less than 2% of the annual penta-BDE consumption occurs in Europe and Asia (UNEP, 2010b). Octa-BDE usage in North America accounts for more than 70% of the world's total octa-BDE consumption (3-5 kt), with 15-20% of the total consumption occurring in Europe (Abbasi et al., 2019). In 1992, deca-BDE was mostly used in North America (40%), followed by Japan (25%) and Western Europe (25%), while deca-BDE consumption in Asia (except Japan) significantly increased to 40% of the global market demand by 2000, with 14% and 44% of the global market demand in the early 2000s allocated to Europe and North America, respectively (Figure 2). Despite the cease in deca-BDE production in Europe in 1999, deca-BDE consumption remained constant within the range of 7 and 12 kt from the mid-1980s to 2010 (Earnshaw et al., 2013). Due to the harmful characteristics of PBDEs that were well known to environmental authorities in the early 2000s, the production of commercial penta- and octa-BDE mixtures ended in 2004 in the European Union and U.S., except for deca-BDE mixtures, which ended in 2014 in North America. Pentaand octa-BDEs were further listed in Annex A of the Stockholm Convention (SC) in 2009, while deca-BDE was added in 2017.

Technical PBDEs with high environmental persistence are resistant to physical, chemical and biological degradation. PBDE congeners with bromine atoms in ortho-substituted positions to the ether bond possess higher vapor pressure (Wong et al., 2001). As typical hydrophobic contaminants, the logarithmic octanol-water partition coefficients (log*K*_{OW}) of PBDEs vary from 4.3 to 9.9 (Table A1), hence enabling their bioaccumulation in organisms as well as enabling significant binding to aerosol, sediment, and soil particles, especially for highly brominated compounds (Cetin et al., 2019; Harner and Shoeib 2002). In contrast, lower brominated PBDEs tend to be unbound in the environment in the forms of being dissolved and gaseous and are more soluble in water. These results in lower brominated PBDEs with a greater tendency to volatilize from the Earth's surface, and thus, these forms can be transported via air from their source areas (Harner and Shoeib 2002).



Figure 2: Global and regional production of PBDEs (kt) from 1970 to 2020

Source: Abbasi et al. 2019.

2.2 Sources and long-range environmental transport

Environmental PBDEs are point and/or nonpoint sourced. The former includes products from the incineration of PBDE-containing waste, PBDE-containing products on fire, release from inuse PBDE-containing products, PBDE incorporation into polymers, recycling of PBDE-containing plastic products, and textile finishing processes with PBDEs (Alcock et al., 2003). Additional point sources of PBDEs in the environment are hospital waste and municipal or hazardous waste incinerators, leachates from waste disposal, and effluents from factories producing flame-retardant polymers (Clarke et al., 2008). Atmospheric transport and deposition, e-waste importation and disposal, and the application of PBDE-incorporated products (e.g., furniture, plastic, and electronics) constitute typical nonpoint sources (Agrell et al., 2004; Ni and Zeng 2009). For high-latitude polar regions farther from PBDE sources, atmospheric transport and deposition are more responsible for their presence (Hung et al., 2010).

Point-sourced PBDEs can diffuse to neighboring environments and cause contamination, which is termed the 'halo effect'. Additionally, PBDEs are well-known pollutants that undergo long-range atmospheric transport (LRAT) after volatilization from their sources. Based on shipborne measurements combined with the Lagrangian particle dispersion model FLEXPART, Mulder et al. found that atmospheric PBDEs in the central and eastern Mediterranean were sourced from the Izmir-Aliaga area via LRAT (Mulder et al., 2015). The long-range transport potential of PBDEs to remote areas has been evaluated by Wania and Dugani (Wania and Dugani 2003) based on a variety of multimedia fate and transport models, namely, the TaPL3-2.10, ELPOS-1.1.1, Chemrange-2, and GloboPOP-1.1 models. Comparable predictions from these four models were yielded, showing that only a small proportion of PBDEs residing in air and water could migrate over long-range distances, with a very low potential for the highly brominated congeners to reach remote areas in comparison with the lighter ones. These modeling results were consistent with field measurements in remote polar regions, where the lighter components

of commercially produced PBDE mixtures were predominant in air (Hung et al., 2016; Ma et al., 2019). Apart from LRAT, PBDE transport associated with water masses was investigated in pan-Arctic shelf seas and the interior basin, resulting in PBDE emissions in a minor fraction ultimately being long-range transported to the Arctic Ocean through PBDE accumulation assessments (Salvado et al., 2016).

Environmental photodegradation is a prominent abiotic process for PBDE transformation into lower brominated congeners and derivatives that may be associated with more toxic and bioaccumulative potential (Kajiwara et al., 2008; Zhao et al., 2015). PBDEs can experience direct photodegradation via exposure to sunlight, with heavier congeners being apt to absorb longwavelength ultraviolet (UV) light (Zeng et al., 2008). Indirect photolysis of PBDEs is triggered by environmental photosensitizers (e.g., organic matter and ferric ions) through light absorption, the production of reactive oxygen species, and subsequent reactions with PBDEs (Zhao et al., 2015). PBDE photodegradation in aquatic environments largely depends on chemical species, including humic substances, metal ions, and halide ions (Leal et al., 2013; Zhao et al., 2014), while gas-phase PBDEs are subject to direct photolysis and photooxidation by OH radicals (Raff and Hites 2006). In the gaseous environment, direct photolysis can significantly remove PBDEs with more than three bromine atoms, while congeners with one or two bromine atoms react preferentially with OH radicals (Raff and Hites 2007). Regarding particle-bound PBDEs, their photolysis rate strongly affects particle geochemistry, such as porosity, water content, organic carbon content, and the presence of metal oxides (Hua et al., 2003; Soderstrom et al., 2004). The photodegradation pathways for particle-associated PBDEs include stepwise reductive debromination, which leads to lower brominated congeners, as well as intramolecular HBr elimination on PBDEs for cyclization with the formation of dibenzofuran (Kajiwara et al., 2008). Additionally, oxidative and reductive debromination can occur upon the photoexcitation of TiO₂ (Park et al., 2013).

For biodegradation under anaerobic conditions, reductive debromination, including ortho-, meta- and para-debromination, dominates the PBDE removal pathways. The half-lives of PBDE debromination in sediment and sludge under anaerobic conditions, following first-order kinetics, varied from 2.67 d (BDE-3) to longer than a decade (BDE-209), ultimately indicating a slow process of debromination under anaerobic conditions (Shih et al., 2012; Tokarz et al., 2008), and general debromination difficulty for heavier PBDEs with lower bioavailability. PBDE degradation processes under aerobic conditions involve adsorption on the cell surface, assimilation into cells, breakdown of the aromatic ring, and subsequent mineralization (Wang et al., 2016). The degradation rates of heavier PBDEs are lower than those of lighter PBDEs under aerobic conditions, and PBDE degradation rates under aerobic conditions are basically higher than those under anaerobic conditions.

2.3 Concentrations in environmental matrices

2.3.1 Air and rainwater

Atmospheric PBDE levels from station- and ship-based measurements in polar regions are summarized in Table 1. Air monitoring under the AMAP shows that PBDE concentrations at most stations were declining, apart from Stórhöfði, Iceland, where BDE-99 and BDE-100 each increased by a small margin. This result differed from the seemingly stable trend from 2002 to 2012 observed at Alert (Hung et al., 2016), which may respond to effective regulation and hence a reduction in technical penta- and octa-BDE releases from North America and Europe in the mid-2000s. Some relatively more volatile PBDE congeners (e.g., BDE-47, 99, and 100) exhibited a strong cyclical pattern at Alert and Pallas, with a maximum value observed in summer, when

the re-volatilization of these congeners likely controlled the atmospheric levels. However, the temporal trend of the air BDE-209 level in the Arctic is still contested; BDE-209, predominating in the BDE pollution profile in the recent Arctic, was moderately decreased at Alert, but this result was inconsistent with its slow or non-changing tendency at Zeppelin and Andøya (Wong et al., 2021). Through an air survey of PBDEs in West Antarctic from 2011 to 2017, no clear temporal trend was observed, with BDE-28 and BDE-47 dominating the PBDE air profile as well as a high detection rate of BDE-183 (Hao et al., 2019a). This result indicates a necessity for continuous attention to PBDE emissions from both environmental reservoirs and unintentional sources.

To our knowledge, PBDE data on wet deposition in the high Arctic/Antarctic are not available, with limited reports in circumpolar countries. ter Schure and Larsson investigated 9 PBDEs in precipitation in southern Sweden, with a mean total concentration of 209 pg/L and an indication of efficient scavenging for particle-bound PBDEs in air by wet deposition (ter Schure and Larsson 2002).

Location	Sampling year	BDE-47	BDE-99	BDE-100	BDE-154	BDE-183	ΣPBDE
West Antarctic	2017-2018	0.01-0.1	nd-0.1	nd-0.04	nd-0.03	nd-0.2	0.3-0.9
West Antarctic	2015-2017	0.03-0.05	nd-0.1	nd-0.04	nd-0.2	0.1-0.5	0.5-1.5
King George Island	2009-2010	0.09-0.20	0.01-0.06	nd	0.01-0.10	0.1-0.69	0.67-3.0
King George Island	2011-2014						0.60-16
Canadian High Arctic	2002-2004	0.21-18	0.19-22	0.031-2.5	0.015-0.93	0.018-2.7	0.78–48
European Arctic Site	2012-2013	0.23-4.1	0.07-6.8				1.0-31
Atlantic Ocean	2008	0.34-3.6	0.42-1.3	nd-0.34	nd-0.52	nd-0.28	0.86-6.4

Table 1:	Occurrences of atmospheric PBDEs	(pg/m	³) in Polar	Regions	(mean and	average)
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nd = not detectable

Source: Hao et al. 2019, Li et al. 2012, Wang et al. 2017, Su et al. 2007, Salamova et al. 2014a, Li et al. 2011.

2.3.2 Snow and ice core

Snow cover acts as a temporary storage reservoir, particularly in high-latitude areas (Table 2), and it significantly affects the fate of organic chemicals. Snowmelt concentrations of PBDEs in the 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, wherever detectable, in the Antarctic (Khairy et al., 2016). Due to the relatively lagged involvement of deca-BDE in the SC and the proximity of the Arctic to deca-BDE application areas, the three-year snow pit collections from the Devon Ice Cap showed a predominance of BDE-209 with concentrations of 680–100,000

pg/L, indicating no clear temporal trend deposition fluxes of BDE-209 (Meyer et al., 2012). Because of low geographical variability, snow could not provide information about the spatial evolution of PBDEs in larger sections of polar regions.

Based on ice core segments, the historical development of PBDE pollution was studied in polar regions (Table A2). PBDE presence in ice cores of the Arctic was first revealed by Lacorte et al. in 2007 (Lacorte et al., 2009), with only BDE-47 and BDE-99 detected at the low pg/L level. The PBDE deposition history reconstructed from 1953 to 2005 in an ice core from Holtedahlfonna, Svalbard, reflected high amplitude variability of BDE-209 inputs with time, with its levels ranging from 0.33 to 6.95 ng/L, suggesting there was LRAT of BDE-209 to the Arctic during haze periods (Hermanson et al., 2010).

PBDE (pg/L)	Vegetation Island	David Glacier	Mid poin	GV5 Itase	Faraglione camp	Palmer Station— Ross Sea
BDE-17	IDL	1.5	0.9	0.6	nd	
BDE-28	1.6	4.4	1.8	1.7	2.1	
BDE-71	0.6	nd	nd	nd	12	
BDE-47	96	170	59	52	66	
BDE-66	1.6	2	0.9	0.7	0.7	
BDE-100	7.2	19	nd	nd	nd	
BDE-99	120	120	48	180	210	
BDE-85	8.3	8.6	8.8	27	17	
BDE-154	5.9	5.1	2.8	nd	nd	
BDE-153	6.6	5.5	3.9	3.8	2.3	
BDE-183	nd	nd	nd	nd	nd	
ΣPBDE	240	340	130	260	310	49–470

Table 2:Concentrations of PBDEs (pg/L) in snow in areas of high latitude.

nd = not detectable

Source: Vecchiato et al. 2015a, Dickhut et al. 2012.

2.3.3 Seawater, river water, and lake water

The PBDE contents in water from marine and terrestrial environments of polar regions are provided in Table 3 and A3, in which only a few studies have addressed PBDE occurrences in rivers and lakes. The PBDEs present in the Ob and Yenisey Rivers in the Russian Arctic tended to be highly associated with particulate matter in water, with only PBDE-37, 47, and 99 exceeding the level of 1 pg/L (Carroll et al., 2008). The dissolved total PBDEs (12 congeners) in three lakes from the Canadian Arctic ranged from n.d. to 21.4 pg/L, with the highest detection frequency observed for BDE-47 (Table A3); in contrast, the other congeners were detected only intermittently (McDonough et al., 2018). PBDE-bearing seawater has been abundantly studied (Table 3). Previous research in the East Greenland Sea and along a transect from East Asia to the Arctic found BDE-47 and 99 to be dominant, with concentrations less than 1 pg/L (Moeller et al.,

2011a; Moeller et al., 2011b). PBDEs in the polar mixed layer from throughout the Arctic Ocean ranged from 0.3 to 11 pg/L, with higher concentrations found in the pan-Arctic shelf seas (Salvado et al., 2016).

Location	Sampling year	PBDE species	BDE-209	PBDE
Chukchi Sea	2005	14	0.1-1.5	1.1-2.4
East Siberian Sea	2008	14	0.6–0.9	0.9-1.0
Laptev Sea	2008	14	0.6-5.5	0.6-5.5
Kara Sea	2008	14	0.7	1.3
Barents Sea	2001	14	0.6-7.8	0.9-8.3
Norwegian Sea	2001	14	1.5-10	1.9–11
Beaufort Sea	2005	14	2.1-4.1	3.9–5.9
Central Arctic Ocean Basin	2001	14	0.2-3.4	0.2-4.2
East of Greenland	2005	14	0.3	0.9–1.5
East of Greenland	2009	10	-	0.005-0.64
Chukchi Sea and East of Asia	2010	10	nd-0.2	nd-0.8
Kara Sea	2003	43	-	1.8-11

Table 3:	Concentrations of PBDEs	(ng/l) in	seawater in	nolar regions
Table 5.	CONCENTRATIONS OF F DDLS	(PS/L) III	i seawatei iii	pulai regiuns

nd = not detectable

Source: Salvado et al. 2016, Moeller et al. 2011a, Moeller et al. 2011b, Carroll et al. 2008.

2.3.4 Sediment and soil

PBDEs have been regularly investigated in soils and sediments of Antarctic and remote northern polar regions (Tables A4 and A5). In addition to LRAT-driven PBDE enrichments in soils of polar regions, PBDE accumulations related to release from research stations, where soil PBDE levels were even comparable to industrial and e-waste regions, are of concern. Vecchiato et al. found soil PBDEs at levels from 0.77 to 33 ng/g (sum of 13 PBDEs) at the Italian Zucchelli station, Antarctic, which were comparable to measurements from industrial and e-waste regions (Vecchiato et al., 2015b). PBDEs in surface sediments have been monitored in Arctic/Antarctic marine and terrestrial environments, including the Bering Sea, Chukchi Sea, Canada Basin, remote lakes in the Norwegian Arctic, and Victoria Land in East Antarctic (Cai et al., 2012a; Corsolini et al., 2019; Jiao et al., 2009). The PBDEs in these sediment samples varied from n.d. to hundreds of nanograms per gram, with their compositional profiles differing spatially.

2.3.5 Biota

PBDE enrichments in biota are provided in Table A6. Studies on PBDE in biotic samples, including phytoplankton, krill, fish, and fur seal milk, from the Antarctic 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). Taking PBDEs in fur seal milk as an example (Figure 3), the average total PBDE concentration (of 7 PBDEs) significantly increased by ca. 4 fold. This result contrasts with a general decreasing trend in the Northern Hemisphere. A time-series investigation of marine mammals, seabirds, marine and freshwater fish and blue mussels in the context of the AMAP showed a typical trend of increasing concentrations up to the mid-2000s for BDE-47, followed by a decrease (Riget et al., 2019). However, the 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).





Source: Markham et al. 2018.

2.4 Environmental pathways

The air-sea gas exchange and dry particle-bound deposition of PBDEs have been studied previously in polar oceans, such as the East Greenland Sea and North Pacific, along high Arctic transects, where both air-sea gas transfer and dry particle-bound deposition contribute to PBDE inputs into the sea (Moeller et al., 2011a; Moeller et al., 2011b). Studies have demonstrated the biotransformation of higher brominated PBDEs to lower brominated congeners, generally correlated with high levels of bioaccumulation (Riu et al., 2008). The analyses of trophic magnification factors and species-specific bioaccumulation factors of PBDEs in the Canadian Arctic marine food web suggest that PBDEs are absorbed by homeotherms and accumulate rather than biomagnify, 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 for PBDEs in the food web of the Ross Sea, Antarctic (Corsolini et al., 2017). Snowpack plays a special role in the POP cycle in snow-covered regions, as it strongly influences their air burden. However, limited data on the air-snow exchange of gaseous PBDEs in the western Antarctic Peninsula indicate that atmospheric deposition functions as a major source of PBDEs in snow (Khairy et al., 2016).

Studies have revealed that environmental PBDEs adversely impact thyroid hormones, endocrine systems, and neurobehavioral development, and they also induce cancer (Herbstman et al., 2010). The ecosystem toxicity of PBDEs in polar regions has been assessed in laboratory animals captured from the field. After exposure of emerald rock cod (an Antarctic fish) to PBDEs, obvious physiological changes such as total lipid, lipid peroxide, and protein carbonyl concentrations in emerald rock cod liver were observed (Ghosh et al., 2013). Risk assessments of human exposure via ingestion to indoor dust-associated PBDEs from Antarctic research stations overall showed no adverse effect on human health (Corsolini et al., 2021). Based on a recent study on PBDE distributions in Antarctic benthic invertebrates collected from local pollution source-free sites (Krasnobaev et al., 2020), BDE-209 was found to be substantially enriched in five targeted invertebrates. This result contrasts with PBDE congener profiles in Antarctic air that were previously obtained from monitoring stations, evidencing the source role of LRAT and deposition for BDE-209 in the Antarctic ecosystem.

3 Polycyclic aromatic hydrocarbons (PAHs)

3.1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are composed of at least two fused aromatic rings. Derivative compounds such as nitro-, oxy-, and hydroxy-PAHs, which are receiving increasing recognition as a result of their significant health and environmental hazards, are transformed PAH products that are derived from incomplete combustion processes or formed as products from atmospheric oxidant-initiated reactions (Zhang et al., 2011). Due to their semi-volatility, relatively higher octanol-water partition coefficients, and LART potential (Cabrerizo et al., 2014; Ma et al., 2010). PAHs can be globally redistributed and ultimately deposited in polar regions. Once deposited, PAHs are subject to partitioning, exchange, and diffusion processes, leading to their ubiquity in air, water, soil, sediment, and biota (Deelaman et al., 2021; Pouch et al., 2021; Yu et al., 2019).

The known carcinogenic and mutagenic effects of PAHs on biota and humans along with their anthropogenic releases and persistence drive PAHs deemed POPs by the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (LRTAP) to minimize their release. Afterward, PAHs were include in the Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR) (OSPAR, 2000; UNECE, 1998). In the mid-1970s, 16 parent PAHs were prioritized by the U.S. Environmental Protection Agency (U.S. EPA) for emission control based on their toxicity, analysis ability and environmental occurrence, and this number was further extended to include 18 parent PAHs and 16 groups of prominent C1- to C4-alkyl PAH derivatives.

The global atmospheric emission inventory of 16 U.S. EPA priority PAHs estimated by Zhang and Tao (2009) in 2004 was 520 Gg/year, with major sources from biofuel, wildfire, and consumer product usage. China had the highest PAH emissions (114 Gg/year), followed by India and the United States (Zhang and Tao 2009). Based on the PKU-FUEL-2007 dataset, the global atmospheric emissions of the 16 U.S. EPA priority PAHs from 69 major sources were estimated in 2007, with the highest PAH emission densities observed in South Asia (87 Gg), East Asia (111 Gg), and Southeast Asia (52 Gg), collectively contributing half of the global total PAH emissions (Shen et al., 2013). Additionally, the time series simulation for PAH emissions from 1960 to 2030 indicated a decreasing global trend. Considering PAH emissions from circumpolar countries in 2007 by Shen et al. (2013), emitted PAHs were relatively low, contributing less than 10% to the total global emissions (Shen et al., 2013).

PAHs commonly possess low vapor pressures and water solubility and have high boiling and melting points, with an enhanced tendency of these characteristics for heavier PAHs (Table A7). As semi-volatile compounds, PAHs with vapor pressures generally in the range of 10^{-6} and 10^{-2} Pa can partition between gas and particle phases and are readily long-range transported to regions far from source areas via air. Due to high octanol-water/air partition coefficients, PAHs are particle reactive contaminants that tend to associate with organic matter (e.g. black carbon and coal char) by diffusion through and sorption onto particle surfaces in both aqueous and atmospheric environments (Fernandez et al., 2002; Mitra and Dickhut 1999). Based on modeling, PAHs in air were less persistent, with half-lives ranging from several hours to days, than long-lived sediment or soil-associated PAHs (with half-lives of ca. decades) (Lammel et al., 2009). Air PAHs experience wet and dry scavenging, and the latter significantly affects their residence times in air, particularly for heavier PAHs (\geq five rings) in the aerosol phase, while chemical reactions with OH radicals, NO₃ radicals, and ozone represent a main sinks for gasphase PAHs (Keyte et al., 2013).
3.2 Sources and long-range environmental transport

PAHs are primarily formed by the incomplete combustion of carbon-containing materials, including wood, coal, and diesel, thus possessing both point (e.g., oil spill) and nonpoint (e.g., vehicle emissions) sources (Yunker et al., 2002). Based on core profiles of PAH pollution extracted from aqueous environments worldwide (Engels et al., 2018; Fernandez et al., 2000; Liu et al., 2012), PAH environmental inputs have closely followed socioeconomic development, as indicated by fairly excellent correlations between PAH sedimentary fluxes and energy consumption and pollution density. This process renders areas with intensive anthropogenic activities as the main emitters of PAHs. PAH derivatives are derived from primary combustion sources as well as atmospheric secondary formation (Atkinson and Arey 1994). As with the source apportionment results in multiple media from low-latitude areas (Albinet et al., 2008; Alves et al., 2017; Lin et al., 2015), nitrated, hydroxylated, and oxygenated PAHs were primarily sourced from combustion-related emitters based on few available reports in polar regions (Drotikova et al., 2020; Drotikova et al., 2021).

The semi-volatile properties, slow photochemical degradation, and limited atmospheric deposition allow PAHs to undergo LART to polar regions, accompanied by multi-hopping enroute. The long-range transport of air PAHs and derivatives has been evidenced by continuous monitoring from remote sites in the Arctic and/or modeling simulation. The PAH concentrations at these remote sites are one order of magnitude lower or even higher than the concentrations in source regions (Hung et al., 2005). However, to date, the long-term monitoring of PAHs and derivatives in Antarctic air has rarely been performed. Sofowote et al. applied combined positive matrix factorization and potential source contribution function analysis to assess PAH transport to a sub-Arctic site and further identify source contributors of geographically dispersed PAHs (Sofowote et al., 2011). The PAHs in the observed sites therein had undergone both relatively short and long distances from North American and Asian sources. Friedman and Selin used the global 3-D chemical transport model GEOS-Chem to simulate seasonal LRAT of PAHs to the Arctic with considerations of effects from atmospheric partitioning, deposition, and pyrolysis on LRAT (Friedman and Selin 2012). The results showed that air PAHs in the Arctic exhibited obvious seasonality, with more PAHs enriched in winter. Source contributions to the Arctic transport of PAHs were allocated, with Europe (47-70%), Russia (13-29%), and North America (9-15%) being the top three emitters. To better understand the impacts of climate change and emissions on air PAHs in the Arctic, Friedman et al. furthered the GEOS-Chem-based simulation by coupling it with meteorology from a general circulation model and projected the results for 2050, indicating that atmospheric PAHs in the Arctic would decline by a small margin (ca. 8%) (Friedman et al., 2014). PAHs in seawater have the potential for long-range transport to polar regions via oceanic currents. Cai et al. conducted a survey of seawater PAHs from the western Pacific to the Southern Ocean with sampling transects spanning over ca. 87° latitudes, implying that water masses and phytoplankton significantly influence PAH distribution in seawater (Cai et al., 2016).

PAHs are reactive toward various atmospheric oxidants, such as hydroxyl radicals, nitrate radicals, ozone, and nitrogen dioxide, leading to the formation of nitro-, oxy-, and OH-PAHs as well as other oxygenated species (Atkinson and Arey 2007). Gaseous PAH reactions with OH radicals can proceed through two possible pathways, including (1) OH radical interactions with substituent groups (e.g., C–H groups) and (2) OH addition to the aromatic ring (Reisen and Arey 2002), the importance of which depends on temperature and pressure. PAHs can react with mixtures of N_2O_5 – NO_3 – NO_2 , involving the addition of NO_3 to the aromatic ring, except that NO_3 radicals interact with the substituent groups of PAHs. The reaction of PAHs involving the more dominant NO_3 addition mechanism followed by the preferential addition of NO_2 is expected to

produce nitro-PAH isomers; however, reactions through NO₃ interaction with substituent groups (H-atom abstraction) are not expected to produce them (Keyte et al., 2013). For PAHs, no loss was observed even during their exposure to relatively high O3 concentrations (ca. 4 ×10¹³ molecules/cm³). Reactions with OH radicals for gaseous PAHs and with ozone for aerosol-phase PAHs can both produce oxy-PAHs (Lammel 2015). Many studies concerning the extent and nature of direct emission sources for oxy- and nitro-PAHs in ambient air have been conducted (Jakober et al., 2008). PAH photodegradation can occur in water, as well, through direct photolysis under solar radiation and indirectly enhanced photolysis by chemical transients such as the hydroxyl radical, singlet oxygen, and photochemically excited states of dissolved organic matter (Jacobs et al., 2008; Miller and Olejnik 2001). Dissolved organic matter can accelerate the photodegradation of lighter PAHs such as phenanthrenes by promoting the formation of reactive intermediates, but inhibit the photodegradation of larger PAHs by binding PAH molecules (Xue et al., 2019). Additionally, salinity and suspended particles exert certain influences on PAH photodegradation in natural water (Shang et al., 2015).

PAH biodegradation occurs under both aerobic and anaerobic conditions, and the biodegradation rate is highly correlated with multitudinous factors, including environmental conditions (e.g., pH, temperature, oxygen), microorganisms (population and type), degree of acclimation, nutrient accessibility, chemical property, and their partitioning characteristics (Haritash and Kaushik 2009). Typical PAH-degrading bacteria are Pseudomonas aeruginosa, Pseudomonas fluoresens, *Mycobacterium spp., Haemophilus spp.*, and *Rhodococcus spp* (Haritash and Kaushik 2009). PAHs in fish are rapidly metabolized to more polar metabolites by triggering the induction of cytochrome P450 family 1A (CYP1A) proteins via an intracellular aryl hydrocarbon receptor (Tomy et al., 2014). Biodegradation-induced variations in PAH profiles in bulk sediment, soil, and sea ice from polar regions have been well documented (Mikael et al., 2003).

3.3 Occurrences in environmental matrices

3.3.1 Air and rainwater

Atmospheric PAHs in the polar regions have been investigated by either long-term site-based observations or ship-based expedition during various sampling periods (Table 4); the stations of Alert, Zeppelin, and Pallas were covered by the AMAP for Arctic PAH monitoring since 1992, 1994, and 1996, respectively (Kong et al., 2014). Based on the station Alert survey in Canada from 1993 to 2002, 16 U.S. EPA priority PAHs had values ranging from 110 to 520 pg/m³ and from 38 to 390 pg/m³ in the gas and aerosol phases, respectively, with an overall decline from 1992 to 1996 and an obvious increase in gaseous PAHs in 1998 (Hung et al., 2005). In recent years, a significant decline in PAH contamination in the Arctic was not observed, as anticipated (Yu et al., 2019), despite a decrease in global PAH emissions (Figure 4). This result possibly suggests global warming-induced volatilization of PAHs as a gradually important source in polar regions. In contrast, available studies regarding long-term PAH monitoring of Antarctic air showed a declining trend from 2013 to 2019 (Na et al., 2020a). PAH derivative pollution in the Arctic boundary layer displayed sixfold and two orders of magnitude higher levels of oxy-PAHs than the parent and nitro-PAHs, respectively, with their highest levels observed in the spring (Drotikova et al., 2021). Due to cold condensation, PAHs and their derivatives in precipitation in the high Arctic/Antarctic regions have seldom been determined, except for some reports from circumpolar regions (Lunde and Bjorseth 1977).

Location	Year	∑PAH (gas phase)	∑PAH (aerosol phase)	∑PAH (gas + aerosol)
Arctic Ocean	2008		7-240	
Barents Sea	2016	20000-22000	1400 (250-3000)	
Kara Sea	2016	19000-22000	200 (230-270)	
Leptev Sea	2016	19000-22000	300 (230-890)	
East Siberian Sea	2016	19000-23000	2100 (240-3300)	
North Pacific to the Arctic Ocean	2012			73
Alert, Canada	1992-2015			340
Zeppelin, Norway	1994-2015			650
Pallas, Finland	2009-2015			700
Arctic Ocean	2010	3400 (2200-4700)	6.6 (0.2-14)	
Svalbard	2018			3.9 (1.4-8.1)
Svalbard	2018			470 (330-740)
Svalbard	2018			37 (30-46)
Alert, Canada	1991		21 (nd-850)ª	
North Pacific to the Arctic Ocean	2014	3.7 (0.67-13)		
North Pacific to the Arctic Ocean	2018	1700 (1400-1800)	47 (28-66)	
Svalbard	2015	38000 (18000-60000)	2100 (1100-7200)	
Tromsoya, Norway	2013-2014	15 (2.6-34)		
Bransfield Strait, Antarctic	2014	2.4- 6.9	0.90-1.8	
Livingston Island, Antarctic	2014-2015	150 (18-870)		
Fildes Peninsula, Antarctic	2013-2019	6500 (360-59000)	640 (17-7200)	
Arctic Ocean	2016	1.7	0.03	

Table 4:	Occurrences of atmospheric PAHs (pg/m ³) in polar regions (mean and range)
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^a nd = not detected

Source: Paatero et al. 2009, Ji et al. 2019, Wang et al. 2013, Yu et al. 2019, Ma et al. 2013, Drotikova et al. 2020, Singh et al. 2017, Zheng et al. 2021, Na et al. 2021a, Na et al. 2021b, Casal et al. 2018a, Cao et al. 2018, Casal et al. 2018b, Na et al. 2020a, Chen et al. 2021.



Figure 4: Temporal trends of mean atmospheric concentrations for Phe, Pyr, and BaP at three Arctic air monitoring sites

Source: Yu et al. 2019

Table 5: Concentrations of PAHs (ng/L) in snow in polar regions (mean and range)

Location	Sampling year	PAH species	∑ран
Northern Victoria Land to the East Antarctic plateau	2011-2012	Parent; 16	32 (0.65-140)
Svalbard, Norway	2017	Parent; 16	55 (2.6-300)
Fildes Peninsula	2009	Parent; 16	120 (52-270)
Dome C, Antarctic	2016	Parent + Alkylate; 17	2.7 (1.6-4.4)
Ekström Ice Shelf, Antarctic	2002-2005	Parent + Alkylate; 10	130 (24-190)
Livingston Island, Antarctic	2014-2015	Parent + Alkylate; 53	4.9 (0.39-9.2)
Victoria Land, Antarctic	2020-2021	Parent; 16	32 (24-39)
King George Island, Antarctic	2017	Parent; 9	0.81 (0.51-1.4)

Source: Vecchiato et al. 2015a, Vecchiato et al. 2018, Na et al. 2011, Xie et al. 2020, Kukučka et al. 2010, Casal et al. 2018a, Arcoleo et al. 2021, Szumińska et al. 2021.

3.3.2 Snow and ice core

A majority of the Arctic/Antarctic environmental studies revealed snow/ice cores as good preservatives for PAHs (Table 5 and 6). PAHs in surface snowmelt from the high Arctic substantially varied spatiality from several to hundreds of nanograms per liter (or in the range of n.d. to thousands of nanograms per gram for snowpack), reflecting the main contributions from LRAT (Lebedev et al., 2018; Masclet et al., 2000; Vecchiato et al., 2018). Additionally, PAH levels of the same order of magnitude were encountered in Antarctic snow (Vecchiato et al., 2015a). However, arising from discrepancies in PAH emitters in the polar regions, the composition profiles of PAHs in surface snow differed (Lebedev et al., 2018; Xie et al., 2020).

Through the retrieval of time-dependent PAH pollution profiles in ice cores from polar regions, some historic epochs responsible for depth variability in PAH levels, in addition to long-range sources from low latitudes, were revealed, such as volcanic eruptions and mining activity (Masclet et al., 2000). The PAH contamination level achieved in these ice cores depends on the proximity to human-dominated sources (Table 6). Ice cores collected from the Antarctic (70°41′S, 158°52′E, 1950 m a.s.l.) were less PAH-contaminated, with PAH levels less than 10 ng/L throughout the core (Giannarelli et al., 2017).

Location	Sampling Year	Deposition period	PAH species	∑ран
Agassiz Ice Cap, Ellesmere Island	1993	1963-1993	Parent; 7	220 (35-660)
Lomonosovfonna, Svalbard	1997	1475-1989	Only Nap	10 (nd-53)ª
Greenland	1989	1539-1989	Parent + Alkylate; 42	580 (1.5-5800)

Table 6: Concentrations of PAHs (ng/L) in ice cores in Polar Regions (mean and range)

^a nd = not detected

Source: Peters et al. 1995, Vehviläinen et al. 2002, Kawamura et al. 1994.

Location	Sampling year	PAH species; number	∑PAH (dissolved phase)	ΣΡΑΗ (particulate phase)
Ross Sea	2000-2001	Parent; 13	1.6 (1.1-4.0)	4.0 (1.5-8.2)
Barents Sea	2016	Parent + Alkylated; 35	70 (12-160)	
Kara Sea	2016	Parent + Alkylated; 35	48 (12-14)	
Laptev Sea	2016	Parent + Alkylated; 35	13 (11-16)	
East Siberian Sea	2016	Parent + Alkylated; 35	40 (11-130)	
North Pacific to the Arctic Ocean	2014	Parent; 11	7.9 (1.8-16)	
Arctic Ocean	2016	Parent; 15	1.9	
Pacific sector of the Arctic Ocean	2018	Parent; 16	0.13 (0.034-0.34)	0.006 (0.003-0.013)
Bransfield Strait, Antarctic	2014	Parent; 15	5.4-34	
Prydz Bay	2010-2011	Parent; 9	2 (nd-6.3)	
Ny-Ålesund, Arctic	2017	Parent; 16	2.7 (0.6-6.3)	
Svalbard, Arctic	2015-2019	Parent; 12	15 (0.43-120)	20 (0.20-310)

Table 7:Concentrations of PAHs (ng/L) in seawater in polar regions (mean and range)

Source: Sericano et al. 2001, Ji et al. 2019, Ma et al. 2013, Zheng et al. 2021, Chen et al. 2021, Na et al. 2020a, Cao et al. 2018, Cai et al. 2016, Vecchiato et al. 2018, Pouch et al. 2021.

3.3.3 Seawater, river water, and lake water

PAHs in seawater in polar regions have been abundantly reported through ship-based sampling campaigns (Table 7). The PAH levels in surface seawater varied in magnitude from picograms to nanograms per liter, depending on the sampling period and location. Depth profiles of seawater PAH concentrations have been studied; the Chukchi Sea, for instance, revealed a general decrease in PAH abundance with depth, indicating PAH depletion in the deep sea (Giannarelli et al., 2017). Previously, the PAH abundance in Arctic rivers mainly focused on the Mackenzie River, the largest North American river flowing into the Arctic (Table A8), with total PAHs in the dissolved and particulate phases being up to several hundreds of nanograms per liter (Yunker et al., 2002). PAHs were detected in remote lakes in the Arctic (Table A9), such as lakes from the northwestern part of Wedel Jarlsberg Land in 2012 (Lehmann-Konera et al., 2020) and Lake Abiskojaure in northern Sweden in 2008 (Torneman et al., 2008). The PAHs in these lake samples ranged from n.d. to more than 1000 ng/L.

Figure 5: Geomean ∑PAH concentrations in the Barents Sea sediments in the 1990s (1992-1998) and 2000s (2001-2005)

Panels a, b, and c represent the sum of naphthalene, phenanthrene, dibenzothiophenes and their C1-C3 alkylsubstituted homologs, acenaphthylene, acenaphtene, fluorene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b+k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, perylene, benzo[ghi]perylene, indeno[1,2,3-cd]pyrene and dibenzo[a,h,]anthracene; sum of PAHs with four- to six-ring hydrocarbons, perylene excluded; and sum of naphthalene, phenanthrene, dibenzothiophenes and their alkyl-substituted

homologs, respectively. Symbols I to V denote Svalbard offshore, northeastern Barents Sea, Bear Island Trough, southwestern Barents Sea, and southeastern Barents Sea, respectively.



Source: Dahle et al. 2009.

3.3.4 Sediment and soil

Sediment PAHs in the bottom boundary layer or cores from the Arctic/Antarctic Ocean are compiled in Table 8, with higher PAH loadings found in coastal areas. In addition to PAHs deposited in the polar regions from long-range sources, riverine inputs of PAHs from areas experiencing anthropogenic disturbances, particularly in the Arctic, contributed to substantial PAHs of pyrogenic origin, as indicative of PAH source apportionment in sediments (Elmquist et

al., 2008). This result was in line with the enhanced inputs of anthropogenic PAHs over time in polar regions. Based on investigations in sediments of the Barents Sea in the 1990s (1992-1998) and 2000s (2001-2005), the PAH levels showed no significant changes with time (Figure 5).

Location	Sampling year	PAH species; number	Σран
Arctic Ocean	1993-1994	Parent	280 (35-760)
Arctic Ocean	1993-1994	Alkylated	740 (57-2500)
Arctic Ocean	2010	Parent; 16	16 (2.0-42)
Norwegian Arctic	2005	Parent; 15	28-1100
Yenisey River	1993	Parent; 18	22-810
Ob River	1993	Parent; 18	93-340
Ob River	2004; 2005	Parent + Alkylated; 18	24
Yenisey River	2004; 2005	Parent + Alkylated; 18	130
Lena River	2004; 2005	Parent + Alkylated; 18	80
Indigirka River	2004; 2005	Parent + Alkylated; 18	85
Kolyma River	2004; 2005	Parent + Alkylated; 18	91
Yukon River	2004; 2005	Parent + Alkylated; 18	85
Mackenzie River	2004; 2005	Parent + Alkylated; 18	450
Bering Sea	2010	Parent; 16	58 (50-65)
Chukchi Sea	2010	Parent; 16	68 (52-91)
Canadian Basin	2010	Parent; 16	93 (28-170)
Bering Sea and western Arctic	2014; 2016	Parent + Alkylated; 27	290 (50-900)
Bering Sea and western Arctic Ocean	2010	Parent; 16	71 (37-150)
North Pacific to the Arctic Ocean	2010	Parent + Alkylated; 18	37 (5.8-78)
Baffin Bay, Eastern Canadian Arctic	2008; 2009	Parent + Alkylated; 66	340-2700
Prydz Bay, East Antarctic	2013	Parent; 15	18 (13-31)
Collins Bay, Antarctic	2015	Parent + Alkylated; 36	140 (22-750)
Potter Cove, Antarctic	2010-2011	Parent + Alkylated; 37	34 (12-210)
Maxwell Bay, Antarctic	2010-2012	Parent + Alkylated; 24	40 (4.6-150)

Table 8:	Concentrations of PAHs	(ng/g) in	sediments in Po	lar Regions (mean and range)
		···· / 6/ ····	Sedimento in ro		

Source: Yunker et al. 2011, Dong et al. 2015, Jiao et al. 2009, Sericano et al. 2001, Elmquist et al. 2008, Chen et al. 2018, Lin et al. 2020c, Zhao et al. 2016, Ma et al. 2017, Foster et al. 2015, Xue et al. 2016, Rodríguez et al. 2018, Dauner et al. 2015, Vodopivez et al. 2021.

The PAH levels in terrestrial soils at the Arctic/Antarctic sites are shown in Table A10. As soil PAHs mainly occur through atmospheric scavenging-driven particle deposition, PAH-bearing soils in polar regions also feature anthropogenic impacts, thereby resulting in PAH levels up to dozens of grams per gram observed (Marques et al., 2017). Generally, soils in the Antarctic that are relatively more isolated from direct human influence had less PAH contamination than Arctic soil.

3.3.5 Biota

PAH bioaccumulation in organisms has received extensive attention in the polar region in recent decades. OH-PAHs (PAH metabolites) are considered to be excellent chemical proxies for exposure to the parent PAHs in fish. In polar areas, PAH occurrences in multiple species were reported, such as in fish, invertebrates, penguins, seabirds (including eggs), and mussels, but reports excluded marine mammals to our knowledge (Almeda et al., 2021; Montone et al., 2016; Szczybelski et al., 2016) from which it was noted that invertebrates at low trophic positions accumulated relatively more parent PAHs than did their predators at higher trophic positions.

3.4 Environmental pathways

The PAH exchange tendency between air and seawater/snow has been examined in both the Arctic and the Antarctic based on fugacity modeling. PAH outgassing from and absorption into seawater was found in the Arctic Ocean (Ma et al., 2013; Zheng et al., 2021), independent of sampling period. This result highlights climate change-related ice retreat and thus PAHs escape toward the sea. In contrast, Antarctic environments still played a sink role for long-range transported PAHs (Cao et al., 2018). With respect to air-snow exchange for PAHs in the Antarctic, both a near equilibrium state and net volatilization existed (Cabrerizo et al., 2014; Casal et al., 2018), supporting the secondary source role of snow for PAHs. 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). Through ship-based investigations, the atmospheric dry deposition of PAHs was estimated in the polar regions based on the product of aerosol-phase PAH contents and the deposition velocity of dry particles, with the dry deposition fluxes being significantly lower than the air-sea gas exchange fluxes in both the Arctic and the Antarctic Oceans (Cao et al., 2018; Ma et al., 2013). Despite the existence of PAH bioaccumulation in invertebrates, fish, and seabirds in polar regions (Perugini et al., 2007) this was not true for marine mammals due to trophic dilution effects. The toxicity, persistence, and mobility of polar PAHs in the environment are of global concern. The impacts on the health of northern indigenous communities has been studied by some researchers (Van Oostdam et al., 1999) and more studies on the susceptibility of the food web to PAH contamination have been conducted (Fahd et al., 2020). Another risk to Arctic animal health linked with PAH contamination is the abduction of important cellular components such as proteins, lipids, carbohydrates, or DNA.

4 Organochlorine pesticides (OCPs)

4.1 Introduction

Organochlorine pesticides (OCPs) have been extensively used since 1940s. Most organopesticides are characterized as persistent compounds with half-lives of decades in the environment. Furthermore, they bioaccumulate through the food chain and are transported over long distances from source regions, as evidenced by their accumulation in Arctic wildlife (Grung et al., 2015). Twelve organic compounds were listed as POPs of SC and they are known as the dirty dozen because of their persistence, bioaccumulation, toxicity, and long-range transport (LRT) potential. Among the 12 POPs, aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorocyclohexane (HCH), mirex, and toxaphene are organochlorine pesticides (OCPs), and their usage should be eliminated or reduced worldwide.

OCPs have been widely used worldwide, some of which were banned approximately 40 years ago due to their persistence, LRT potential and high toxicity. However, several OCPs are still in use in some developing countries. By reviewing many studies, the global use/production quantities were estimated and are listed in Table 9. Some estimated data obtained from different researchers have remarkable divergence based on the different models they used. It is estimated that the total production of DDT was approximately 4.5 Mt from the 1940s to 2005 (Li and Macdonald 2005), about one-fold higher than the estimation by Voldner (Voldner and Li 1995). This difference is quite plausible because some DDTs are still used in many countries, such as in manufacturing dicofol. DDT production was estimated to be 4450 t in 2003, 4740 t in 2005, and 6300 t in 2007 in India alone (Corsolini et al., 2011). Among the OCPs listed in Table A11, technical HCHs are the primary OCPs in the world. It is believed that HCH production/use would continue to elevate due to lindane usage, which was included in the second list of POPs and was recently banned.

4.2 Sources and long-range environmental transport

OCPs are a class of human-made chemicals, so the OCPs in the pristine polar areas were input from OCP production and usage areas via LRT routines. Therefore, the OCPs released from the European-Asian and North American continents in the Northern Hemisphere as well from the African and the Australian continents have been confirmed to be the primary sources of OCPs in polar areas by some researchers. Recently, many researchers have paid more attention to OCP fate and transfer from multiple media. OCPs deposited in polar areas can be released again into air via sea-air and ice-water-air exchange, making them secondary sources.

The ratio of OCP isomers, also called the fingerprint, is frequently used to determine the sources of industrial products and input status. HCHs in the environment have two sources: technical HCHs and lindane. The ratio of α -HCH to γ -HCH is an indicator of historical usage, newly technical HCHs or lindane input. The ratios of α -HCH to γ -HCH are markedly different between the Arctic and Antarctic sites (10.2 and 2.5, respectively), suggesting that the possible sources are different (Nash et al., 2017). According to the source apportionment using isomer ratios, the northern temperate and boreal zones are sources of α -HCH to the Arctic. However, the HCH isomer ratios indicated that lindane was recently used in the Southern Hemisphere, resulting in a higher γ -HCH proportion in Antarctic Ocean seawater (He 2013).

Compounds	Persistent and toxic characters	Produce and usage
DDTs	Toxic to human and wildlife, remaining in soil about 10 - 15 years	Global consumption of 0.3 million tons, being banned in 65 countries and restricted production and usage in 26 countries
Aldrin	Carcinogen, lethal dose of 5 g, slowing degrading to Dieldrin in organism and environment	Global consumption of 0.24 million tons, being banned in 72 countries and restricted production and usage in 10 countries
Chlordane	Posing influence on nervous system and immunity system; half-life of 1 year	Global consumption of 0.07 million tons, being banned in 57 countries, and restricted production and usage in 17 countries
Dieldrin	Highly toxic to fish and aquatic animals; half- life of 1 years	Global consumption of 0.24 million tons, being banned in 65 countries, and restricted production and usage in 26 countries
Endrin	Highly toxic to fish, aquatic invertebrates and plants, half-life in soil of 12 years	Being banned in 67 countries and restricted production and usage in 9 countries
Heptachlor	Reproductive and metabolic disorders, possible carcinogen; residue in soil about 2 years	Being banned in 59 countries and restricted production and usage in 11 countries
Mirex	Possible carcinogen, half-life of 10 years	Being banned in 52 countries and restricted production and usage in 10 countries
Toxaphene	Causing thyroid tumors and cancer, residue in soil 12 years	Global consumption of 1.33 million tons, being banned in 57 countries, and restricted production and usage in 12 countries
НСВ	Disturbing immunity and reproductive system; half-life of 2.7 - 2.9 years in soil	Global consumption of 100 - 200 million tons, being banned in 59 countries, and restricted production and usage in 9 countries
HCHs	Half-lives of γ-HCH are 2.3 - 13 days in air, 30 - 300 days in water, 50 days in sediment and 2 years in soil, respectively	Global consumption of 1.40 million tons from 1945 to 1992, being banned in majority of developed countries since 1980s

Table 9.	The toxicities	nroduction and	usage of	several	OCPs in	the world
able 9.	The toxicities,	production and	usage UI	Several		the world

Source: Lin 2007.

The relative proportions of DDT/DDE and DDT/DDD can indicate pollution sources. The high concentrations of Σ DDTs and the high ratio of DDT to DDT+DDD+DDE in the Pechora (Russia) sediment suggest the recent use of DDT in this region (AMAP, 2004). Technical DDT was the dominant source (>94%), which was fresher in Far East Asia than in the North Pacific Ocean and the Arctic. The estimated contribution of dicofol-type DDT was minor. However, o,p'-DDT have a relatively greater contribution from dicofol-type DDT in the North Pacific Ocean and the Arctic (Ding et al., 2009).

Technical chlordane is composed of a mixture with abundant components of TC and CC, which are the current main sources of TC and CC in the environment. In general, a ratio of CC/TC ranging between 1.2 and 1.3 indicates the use of technical chlordane products, while this ratio should gradually decrease in the environment because the degradation of CC might be faster

than that of TC (Tian et al., 2012). Consequently, the historical use or recent input can be determined by the isomer ratio.

The concentrations of OCPs in polar areas were also used to reveal their usage status. For instance, mirex measured in the eastern Antarctic atmosphere might be caused by historical usage in the Southern Hemisphere, while endosulfan-I appeared to show fresh, late-austral-summer input endosulfan-I (Nash et al., 2017).

OCPs can be transformed under natural conditions. DDT can be degraded into DDE via oxidative dehydrochlorination and into DDD via reductive dichlorination. The stability of individual HCHs in marine and atmospheric environments is different and determined by their physicochemical properties. γ -HCH is easily degraded to α -HCH under the action of microorganism metabolism and photochemical reactions (Walker et al., 1999), and α -HCH can transform into β -HCH due to the stable symmetrical structure of the latter (Ya et al., 2017). Based on the identification of the metabolites, transformation of heptachlor was proposed, which was initially metabolized by epoxidation, dechlorination and hydrolysis. Under environmental conditions, parent chlordane compounds are transformed to more stable degradation products, such as oxychlordane and heptachlor *exo*-epoxide(Su et al., 2008).

4.3 Occurrences in environmental matrices

4.3.1 Air

Atmospheric concentrations of OCPs were measured at six Arctic stations: Alert, Kinngait, and Little Fox Lake in Canada, Point Barrow in the USA, Valkarkai in Russia and Zeppelin in Norway on a weekly basis in 2000-2003 (Table A12). The seasonal variations in chlordane-related compounds were weak: monthly averages varied within a factor of 4 among the five stations (excluding Valkarkai) for TC, CC, and trans-nonachlor and less than a factor of 3 for the two degradation products of oxychlordane and heptachlor *exo*-epoxide. Moreover, the temperature effect on chlordane and its degradation compounds was compound-dependent, indicating the combined effects of temperature, secondary emission and degradation. DDTs were relatively lower in the Arctic air. Therefore, the air concentrations of p,p- and o,p-DDD were mostly below the method detection limit (MDL). With respect to dieldrin, the significant negative slope of temperature-dependent relationships suggests that volatilization emissions could contribute to the high summer air concentrations at these sites. Compared to measurements in the 1990s, the air concentrations of dieldrin in 2000-2003 in the Arctic decreased by approximately half. This result is consistent with long-term measurements at Alert, which showed that the half-life of dieldrin in the Arctic atmosphere was approximately 13 years (Hung et al., 2005; Su et al., 2008).

Agricultural chemicals based on the selected analyte repertoire dominated the East Antarctic atmospheric POP profile, with mean concentrations in following order: HCHs > chlordane > endrin > dieldrin > DDTs > mirex. HCH isomers such as α - and γ -HCH were frequently detected in the Antarctic atmosphere, ranging 24 - 1200 and 10 - 530 fg/m³, respectively. HCH isomers play roles that are more prominent in the Arctic atmosphere. The average concentrations of α -HCH in the Arctic were about 2 orders of magnitude higher than those in the Antarctic (Table A12). The ratios of α -HCH to γ -HCH were also markedly different between the Arctic and Antarctic (10.2 and 2.5, respectively), which is consistent with the LRAT pathways and the relative distances between sampling sites to probable sources (Nash et al., 2017).

The total concentrations of the 17 classical OCPs showed an increasing trend from the Sea of Japan to the Bering Sea. It is found that α -HCH and γ -HCH have different latitudinal trends, which might be caused by their physicochemical and fractionation effects during their

northward transport via air masses. The ration of α -HCH to γ -HCH was consistently less than 4, indicating compositional characteristics of a mixture of industrial HCH and lindane (Cai et al., 2010).

Generally, declining trends of atmospheric γ -HCH were observed in Pallas, Alert and Zeppelin throughout the 1990s and early 2000s. For Storhofdi, a declining trend was observed up to 2002, and it was followed by an increase in concentration up to the end of 2003 before the declining trend resumed. The increasing concentration HCHs might be caused by revolatilization during the process of sea ice retreat or the continuous use of pesticides containing HCH.

4.3.2 Seawater, river water and lake water

The emissions of OCPs measured in seawater, lake water and runoff water from polar regions are summarized in Table A13. In the Arctic, the mean concentrations of OCPs in aquatic environment of the Arctic (i.e., sea water, sea ice, and snow) were generally higher than those in the Antarctic. Specifically, the HCH levels measured at the Villum Research Station (81°36'N, 16°40'W) were particularly elevated compared to those observed at Davis Research Station (68°34'S, 77°58'E) toward Anchorage Island in the Antarctic. Three HCH isomers (α -, β -, and γ -HCH) were all detectable in seawater, sea ice, and snow in the Arctic, whereas only γ -HCH was found in these matrices in Antarctic, and α -HCH was only found in seawater. α -HCH was the predominant OCP in Arctic sea water and sea-ice meltwater, ranging 220 - 250 pg/L and 35 - 48 pg/L, respectively.

Dieldrin was a major OCP frequently detected in different environmental matrices in the Arctic and Antarctic. The concentrations of dieldrin in the Arctic are usually 1 or more orders of magnitude higher than those found in the Antarctic. The geographic variations are in agreement with the usage of diedrin and also determined by the short distance between the continental sources to the polar regions. They are also consistent with the earlier studies for OCPs in seawater from both the Arctic and Antarctic (Bigot et al., 2017).

4.3.3 Biota

OCPs have been extensively detected in various organisms in polar regions. Mosses and lichens are the main biotic components of Antarctic terrestrial ecosystems, and they can play an important role as biomonitors of the atmospheric deposition of persistent contaminants (Bargagli, 2008). Significant statistical correlations were found between the reciprocal temperature and log-transformed concentrations of α -HCH, HCB and 4,4-DDE measured in Antarctic soil and mosses. These observations illustrate that temperature is a major factor in determining Earth-scale distribution and accumulation of OCPs, providing additional support for the general validity of describing global distillation effects of OCPs distribution on the Earth (Borghini et al., 2005).

Among the selected POPs measured in Arctic char collected from four lakes (Amituk, Resolute, Char and Hazen Lakes), toxaphene always showed the highest concentrations, followed by $\Sigma DDTs > \Sigma HCHs$. The geometric mean concentrations ranged 1.6 - 170 ng/g ww (wet weight) for toxaphene, 0.54 - 61 ng/g ww for ΣDDT and 0.03 - 4.2 ng/g ww for $\Sigma HCHs$. Most legacy OCPs showed declining trends, which could be attributed to the national or international regulations, bans on their emission and use in circumpolar and neighboring countries. Declining trent of the ΣDDT were observed in all lakes at rates from 7.1%/year to 11%/year. α -HCH also declined significantly in all 4 lakes, with the overall greatest annual change (7.8%/year to 14%/year). Toxaphene was found to have declined significantly only in Amituk Lake (6.4%/year) and Lake Hazen (7%/year), which contrasted with significant increases in Resolute Lake (19%/year) (Ana et al., 2018).

OCPs have high pollution levels in Arctic seabirds and have been identified as a significant stressor for species at risk from environmental change (Miljeteig et al., 2009). DDT, dieldrin and mirex showed significantly declining trends between 1975 and 2003 in three bird species at Prince Leopold Island in the Canadian (Table 10). However, significant increases were observed for Σ HCH in the kittiwakes and fulmars and for β -HCH and *cis*-chlordane in the murres and fulmars (Braune 2007).

Reindeers are long-lived vegetarians that feed on local vegetation, and they are exposed to contaminants mostly through their diet, which is composed of different types of locally grown vegetation, lichen and moss. Therefore, reindeers can be used as valuable bioindicators, serving us to discover the influence of contaminants on polar terrestrial ecosystems. The fur of the Svalbard reindeer was cut to reveal the status of some contaminants. With respect to the 15 studied pesticides, only DDT and its metabolites were above the detection limit, with the highest values found for p,p-DDD, in a single sample from Longyearbyen (57.5 ng/g dw). However, with the exception of this outlier, the p,p-DDD level was much lower in the rest of the samples (median value 1.44 ng/g dw) (Pacyna-Kuchta et al., 2020).

Statistics analysis was performed for the plasma concentrations of major PCBs and OCPs in communities and regions of the Arctic and Subarctic Russian continent. A high exposure population was found in the Chukchi coastal community, which showed primarily relation to the ingestion of marine mammals; however, recent pesticide use is also suspected. Other communities with moderate levels of PCBs had relatively high concentrations of HCB, β -HCH, and p,p'-DDT, and low DDE/DDT ratios (<10), indicating recent pesticide use as well (Sandanger et al., 2009). Relatively high levels of chlordane and p,p-DDE were found in Polar bears from the western Russian Arctic rather than polar bears from eastwards of the region. These results indicated that the western Russian Arctic is highly polluted region in the Arctic and deserves further attention (Lie et al., 2003).

Emerging and legacy POPs were widely detected in biota samples collected from Antarctica, including limpets, amphipods, Antarctic cod, Antarctic icefish, chinstrap penguins and Antarctic skua. Nutrient amplification factors indicate that levels of p,p-DDE, p,p-DDD, cis-chlordane and β -HCH are amplified through the food chian (Kim et al., 2021).

4.4 Environmental pathways

The presence of OCPs in polar areas has been attributed to cold condensation and global fractionation during LRAT (Frank 2003). In winter, there are three southerlies in the Norwegian Sea (40%), Eastern Europe/Siberia (15%), and Bering Sea (25%), accounting approximately 80% of the annual south to north air transport. Therefore, atmospheric transport is the primary pathway in winter. In contrast, the air pressure in summer is relatively higher than that in midhigh latitude regions, making it difficult for air masses to invade the Arctic (Macdonald et al., 2005). It was pointed out that the DDTs and endosulfan in the Arctic were mainly derived from the Europe-Asia continent and were carried by air masses (Ding et al., 2009; Su et al., 2008). Similarly, the DDT distribution in the air of the region in southern latitudes from 23° to 26° was characteristic of a terrestrial source input, identical to the air mass transport trajectory from the African continent to the southwest Atlantic Ocean and Antarctic Ocean (Montone et al., 2005).

Although the concentrations of most OCPs declined in surface water in the western Arctic, the water-air fugacity ratios indicated net volatilization (FR > 1.0) or near equilibrium (FR not significantly different from 1.0), such as those for heptachlor *exo*-epoxide, chlordane and dieldrin. Some of them underwent a process of deposition and volatilization and finally approached equilibrium in the Arctic environment(Jantunen et al., 2015).

Ocean current and river inputs play an important role in OCPs entering the Arctic Ocean, especially from Russian rivers to the estuaries of regional seas and the Arctic basin (Alexeeva et al., 2001; Zhulidov et al., 2000). Deposition, sewage discharge and soil leachate are the main sources of OCPs into rivers and have been well documented for decades. The ocean current and riverine input from the source areas serve as important pathways. For instance, atmospheric LRT was the dominant input pathway for α -HCH entering the Arctic Ocean in 1945 - 1990, while ocean currents superseded the atmosphere to become the major pathway for α -HCH to enter the Arctic Ocean after the early 1990s (Li and Macdonald 2005). With the ban on the use of technical HCH in the 1980s, atmospheric concentrations of alpha-HCH declined and the Arctic Ocean changed from a sink to a source of atmospheric HCH. Archived HCH in cold, snow-covered surface waters of the western Arctic Ocean continues to provide HCH for outflow waters in the Canadian Arctic (Pućko et al., 2013).

Furthermore, some more recent layers of snow and ice melt caused by global climate change have been regarded as possible secondary sources because of the OCPs' re-volatilization and release from glaciers (Bogdal et al., 2009; Khairy et al., 2016; Macdonald et al., 2005). The potential input quantities of OCPs from two glaciers are compared in Table A14. The input fluxes of OCPs from the archipelago were remarkably higher than those of the Agassiz Ice Cap. Similarly, it was observed that snow could be a seasonal secondary source of OCPs in Antarctic (Jones et al., 2005; Potapowicz et al., 2020).

Some animals, including sea birds, whales, pinnipeds, salmons and cods, are usually able to travel a large distance, passing through national boundaries and industrial/agricultural regions with intensive anthropologic activities and arriving in polar areas. Hence, some research inferred that animal migration is another pathway by which OCPs enter polar areas. The grayling in salmon-spawning lakes had twice the concentration of OCPs as those in the salmon-free lakes, and the pollutant composition was similar to that found in salmon. Therefore, in the Alaskan river system, biotransport was found to have a much greater effect on PCB and DDT levels in lake biota than atmospheric input (Ewald et al., 1998). Seabirds can also biotransport certain amounts of OCPs into the Arctic and Antarctic areas, resulting in OCP levels in bird habitats being elevated several fold higher than background concentrations (Blais et al., 2005; Choy et al., 2010; Evenset et al., 2004; Frank 2003; Michelutti et al., 2009). Antarctic fur seals only breed and forage in the Antarctic region in the summer. In the nonbreeding season, they migrate northward to more human-influenced areas, ingesting POPs from these other food webs. Similarly, the migrations of marine mammals have been shown to influence their POP concentrations (Brault et al., 2013).

OCPs eliciting endocrine disruption and immune dysfunction in birds have been well documented by a large amount of laboratory research. Specifically, some OCPs cause delays in laying time and increases in nonviable eggs. In the Antarctic, OCP may delay reproduction and reduce fetal growth in Skuas. However, parental OCP residues were not associated with the occurrence of dead eggs. Moreover, OCPs were negatively related to the egg thickness of the ivory gull *Pagophila eburnean* in Svalbard and the Russian Arctic (Miljeteig et al., 2012). However, even though males had higher OCP levels than females, there was no significant relationship between OCPs and reproductive variables in males, and there was no association between OCPs and adult regression rates among breeding seasons. (Bustnes et al., 2007).

Location	Sampling year	Name	Sample	нсв	HCHs	Chlordane	DDE	DDD	DDTs	Dieldrin	Mirex
Svalbard	1991-1994	polar bears	blood	120	190		58				
Franz Josef Land	1995	polar bears	blood	220	220		84				
Kara Sea	1991, 1994	polar bears	blood	260	300		94				
East-Siberian Sea	1992-1993	polar bears	blood	190	410		38				
Chukchi Sea	1987, 1989-1992	polar bears	blood	240	580		19				
Prince Leopold Island	1975	black-legged kittiwakes	egg	75	4.0	58	240			14	15
Prince Leopold Island	1976	black-legged kittiwakes	egg	89	4.8	82	420			12	26
Prince Leopold Island	1987	black-legged kittiwakes	egg	33	3.6	38	110			8.8	8.6
Prince Leopold Island	1993	black-legged kittiwakes	egg	24	5	33	70			7.5	9.5
Prince Leopold Island	1998	black-legged kittiwakes	egg	25	5.2	32	60			9.2	7.3
Prince Leopold Island	2003	black-legged kittiwakes	egg	16	7.7	45	44			5.3	4.6
Svenskøya	2007	ivory gull Pagophila eburnea	egg	620	110	1800	13000	6.9	230	nd	300
Nagurskoe	2007	ivory gull Pagophila eburnea	egg	930	150	2900	30000	nd	390	560	480
Cape Klyuv	2007	ivory gull Pagophila eburnea	egg	650	140	1600	15000	nd	400	530	350
Domashny	2007	ivory gull Pagophila eburnea	egg	660	290	1200	11000	nd	220	220	220

Table 10:The concentrations of OCPs in biota (ng/g) in the Polar Regions

nd: not detectable

Source: Pacyna-Kuchta et al. 2020; Lie et al. 2003; Braune 2007; Miljeteig et al. 2009; AMAP 2004.

5 Polychlorinated biphenyls (PCBs)

5.1 Introduction

Polychlorinated biphenyls (PCBs) have been used as industrial fluids, flame retardants, diluents, hydraulic fluids, and dielectric fluids. PCBs have a biphenyl ring as their main skeleton with 2 to 9 hydrogen atoms on the rings being substituted with chlorine atoms, as shown in Figure 6. Generally, a total of 209 theoretical PCBs exist, ranging from 3 monochlorobiphenyls to decachlorobiphenyl, and approximately 150 PCBs are found in the environment (Hansen et al., 1999). Some PCB congeners elicit similar toxic and biologic effects as dioxin, and these congeners are referred to as dioxin-like PCBs (dl-PCBs), which are classified according to the chlorines in the ortho position: non-ortho and mono-ortho. The World Health Organization (WHO) has identified 12 toxic dioxin-like congeners (dl-PCBs), including four *non-ortho* PCBs (PCB 77, PCB 81, PCB 126, and PCB 169) and eight *mono-ortho* PCBs (PCB 105, PCB 114, PCB 118, PCB 123, PCB 156, PCB 157, PCB 167, and PCB 189) (Martin et al., 2006b; Yoonki et al., 2014).

Figure 6: The general chemical structure of chlorinated biphenyls



Source: SUST

As a class of persistent and semi-volatile compounds, PCBs can transfer through the solid, liquid, or gaseous phases of environmental compartments, and their fate and transport strongly depend on their physicochemical properties. Therefore, some partition coefficients, such as the organic carbon normalized sorption coefficient (K_{oc}), octanol/water partitioning coefficient (K_{ow}) and octanol/air (K_{OA}) partition coefficient, are well acknowledged as the key parameters used to demonstrate multimedia behavior. Quantitative structure property relationship (QSPR) models have been widely utilized to predict these three coefficients (Hansen et al., 1999; Yuan et al., 2016). By reviewing the literature on K_{ow} values and applying QSARs and experimental measurements, PCBs are a class of hydrophobic organic compounds with K_{ow} values ranging from 4 - 7 (Hansen et al., 1999; Razzaque and Grathwohl 2008; Yuan et al., 2016). Moreover, their K_{OA} values are approximately one to three orders of magnitude higher than those of K_{OW} . The K_{OC} values are significantly associated with PCB sorption in soil, sediment and bio-accessibility, and KOA is vital to demonstrate the transport of PCBs between soil particles and air. Numerous studies have presented a series of empirical equations in which $\log K_{OC}$ is linearly or quadratically related to $\log K_{OW}$ and the fraction of organic matter or is tightly associated with solubility. However, K_{OA} is more likely to be affected by temperature (Komp and McLachlan 1997; Li et al., 2003).

Approximately 1.5 million tons of PCBs have been produced globally in the form of complex mixtures serving as dielectric fluids in transformers, capacitors, and plasticizer agents in paint and rubber

sealants since the 1930s (Bidleman et al., 2010; Breivik et al., 2007). Global production was banned in the 1970s, especially in Europe, where PCBs were banned in open circuit applications from 1979, while they were banned in 1985 in the USA and in 1982 in Chile. It was estimated that 1.3% and 11.8% of the amounts of PCBs historically produced were emitted into the atmosphere by the end of 2005 in default and higher scenarios, respectively. The emissions reached a climax in the 1970s, when PCBs were banned in most countries worldwide. Although the primary emissions remarkably declined, the second sources of re-volatilization slowed the decrease in the percentages of PCB emissions, and a subsequent steep decrease began in 2020 (Breivik et al., 2007). Numerous studies have demonstrated that PCBs are ubiquitous in the ambient environment of the Arctic, including in the atmosphere, water, soil and sediments, as well as in the biota from zooplankton to top predators, such as polar bears, seals and humans. Several regional and international monitoring programs have been conducted to systematically investigate POPs in the polar regions, such as the AMAP, United Nations Economic Commission for Europe, European Monitoring and Evaluation Programme (UNECE-EMEP) and Global Atmospheric Passive Sampling (GAPS) Network.

5.2 Sources and long-range environmental transport

PCBs are introduced to pristine polar areas via several pathways, including gas mass movement (Frank 2003; Hao et al., 2019; Montone et al., 2003; Vecchiato et al., 2015a), ocean currents (Kallenborn et al., 2013), animal, migration birds (Blais et al., 2005; Choy et al., 2010; Eckbo et al., 2019; Michelutti et al., 2009; Warner et al., 2019), fish (Evenset et al., 2004; Ewald et al., 1998), bears (Sonne 2010) and revitalization of oceans and melting glaciers due to climate change (Ademollo et al., 2021; Corsolini et al., 2011; Herbert et al., 2005; Jones et al., 2005; Khairy et al., 2016; Ma et al., 2011). Atmospheric long-range transport, well known as the grasshopper effect, plays a predominant role in PCB legacy in the Arctic and Antarctic areas. It was believed that PCBs in the Arctic were mainly derived from Europe, Asia and Canada (Macdonald et al., 2000; Malanichev et al., 2004; Wu et al., 2011), and PCBs in the Antarctic were input from South Africa and South America. The estimated characteristic travel distance (CTD) of PCBs ranged from 578 km (PCB194) to 7411 km (PCB-52) with the TaPL3 model, while they were in the range of 1484 km (PCB194) to 26908 km (PCB153) with the ELPOS model (Frank, 2003; Frank and Chandrasagar, 2003). PCBs are transported from the lowlatitude zone to the Arctic by multiple hops, i.e., repeated cycles of deposition and re-evaporation, as inferred by the Globo-POP model, demonstrating that the compounds with log K_{OW} values of 5 - 8 (the immediate log K_{OA} ranged from 6.5 to 10) exhibited air-surface exchange. However, chemicals with log K_{OA} > 9.5 could not undergo "multiple hops". They can reach the Arctic with LRAT without being deposited along the way (Frank, 2003).

Several studies have indicated that the chiral signature of PCBs in air varies after long-range travel via oceans, lakes and seas, while scientists have not yet revealed the explicit mechanism. For instance, the enantiomer fraction (*EF*) of PCB136 significantly deviated from the racemic values, indicating a stereo selective depletion of PCB-136 in Antarctic air (Wang et al., 2017). This phenomenon was also reported previously in the equatorial Indian Ocean (Huang et al., 2013) and Arctic areas (Bidleman et al., 2012). It was assumed that the fresh PCBs were discharged into air and that the second sources of air-water or air–soil exchange played an important role in PCB *EF* value variation.

Laboratory experiments have proven that OH radicals in the atmosphere readily oxidize PCBs in the gas phase (Mandalakis et al., 2003). Five years of atmospheric monitoring research showed that some lightly chlorinated PCBs declined significantly in the Arctic atmosphere, and this decline was attributed to OH degradation via reaction with OH radicals. However, scavenging processes, e.g., particulate deposition and snow scavenging, might be more important for heavier congeners in the Arctic (Hung et al., 2001).

Once PCBs enter organisms, some PCB congeners will be metabolized by phase I and II biotransformation enzymes, producing more water-soluble compounds. Cytochrome P450 (CYP) enzymes, known as important phase I enzymes, catalyze the direct insertion of hydroxyl (OH)-groups

into polychlorinated biphenyls (PCBs) or the formation of an arene oxide intermediate, which can be further metabolized to OH-PCBs by epoxide hydroxylase (EH). Furthermore, one of the phase II enzymes, glutathione S-transferase (GST), is involved in the formation of methylsulfone (MeSO₂-PCBs) through the conjugation of glutathione (GSH) and peptidase hydrolysis (Helgason et al., 2010). OH-PCBs and MeSO₂-PCBs have been detected in some Arctic seabirds, such as glaucous gulls, blacklegged kittiwake and northern fulmar chicks, as well as in arctic foxes (Helgason et al., 2010; Nøst et al., 2012; Routti et al., 2016; Verreault et al., 2007; Wu et al., 2014).

The biotransformation of PCBs in biota is characterized by a strong species dependence. Remarkable differences in total MeSO₂-PCB concentrations and composition patterns were found in the seabirds of Arctic areas. Specifically, the levels of $\Sigma_{14/8}$ MeSO₂-PCBs found in the livers of northern fulmar chicks collected from Kongsfjorden (Svalbard, Norway) were higher than those of black-legged kittiwake chicks, which were several orders of magnitude lower than those in the plasma of glaucous gulls from Svalbard (Helgason et al., 2010; Verreault et al., 2005).

Figure 7: Annual mean concentrations of Σ 32PCBs and Σ 7PCBs (pg/m³) in air samples at Zeppelin (Svalbard, Norway), 2001-2020



Source: Bohlin-Nizzetto et al. 2014, 2015, 2016, 2017, 2018, 2019, 2020, 2021.

5.3 Occurrences in environmental matrices

5.3.1 PCBs in air

The PCB concentrations in the air of the Arctic ranged from < LOD to several decades pg/m³, which was much higher than those in Antarctic air. Long-term monitoring results have indicated a generally declining tendency of many legacy POPs in the Arctic, especially in the ambient air (Hung et al., 2010; Hung et al., 2016). Declining temporal trends of atmospheric PCBs (2 - 4% per year) were identified

both along Sweden's west coast and in the sub-Arctic area of northern Finland. Low-chlorinated PCBs will remain in the atmospheric compartment for a long time in Scandinavia (beyond 2030). HCHs and PCB-180 were the first two depleted from the Nordic atmosphere before 2020, and then chlordanes and the rest of the PCBs will be depleted between 2020 and 2025 (Anttila et al., 2016). Consistently, a rough comparison between available data indicated that the concentrations of most POPs in the Antarctic atmosphere have decreased over the last twenty years (Bargagli 2008). Figure 7 shows the annual mean concentrations in 32 most toxic PCB congeners (\S 32PCBs) and 7 PCBs used as indicators in Europe ((\S 7PCBs) from 2001 to 2020 (Bohlin-Nizzetto et al. 2014, 2015, 2016, 2017, 2018, 2019, 2020, 2021). 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 in the years 2017 – 2020.

5.3.2 PCBs in seawater, lake water and river water

PCBs in the aquatic environment of the Arctic areas, including seawater and lake water, were quite similar, ranging from < LOD to several 100s (pg/L). Similarly, PCB concentrations in the Arctic were higher than those in the Antarctic. However, there were remarkable differences in PCB concentrations in snow and ice, covering 3 orders of magnitude. Generally, the PCB concentrations in snow and ice were far higher than those in seawater and lake water; therefore, snow/ice melting was considered a secondary source of PCBs in the Arctic Ocean.

5.3.3 PCBs in sediment and soil

Residual levels of PCBs in the solid media of polar areas ranged from < LOD to several 100s (ng/g); therefore, animal feces and plants accumulated more PCBs. Zhang et al. reported that the total PCB concentrations (\sum_{29} PCBs) determined in the environmental media of soil, sediment, plants, bird guano and reindeer feces were in the range of 2.8 - 11 ng/g, 3.1 - 8.3 ng/g, 22 - 56 ng/g, 35 - 51 ng/g, and 32 - 40 ng/g dry weight (dw), respectively, in Ny-Ålesund, Arctic (Zhang et al., 2014). In the Antarctic, the mean concentrations of Σ PCBs were significantly higher in vegetation (6.9 ± 0.81 ng/g dw) than in organic soils (3.7 ± 0.36 ng/g dw) (Aslam et al., 2019). The average concentrations of PCBs in Antarctic lichens and mosses (range 0.1 - 35 ng/g dw) were generally orders of magnitude higher than those in Antarctic abiotic matrices (Bargagli 2008).

5.3.4 PCBs in organisms

The total PCB concentrations in blubber of ringed seals sampled at Holman did not change significantly between the 1980s and the 1990s (Addison et al., 2005). The PCB concentrations in Baltic fishes decreased by approximately 80%, and these samples were collected across the Finnish coast of the Baltic Sea in 1978-2009 (Airaksinen et al., 2014). The PCB concentrations in the livers of arctic foxes (*Vulpes lagopus*) decreased 4-11% per year. Climate-related changes in Arctic fox diets may affect pollutant concentrations in arctic foxes from Svalbard. The concentration of PCBs in blubber of Antarctic minke whale Balaenoptera bonaerensis males (20-25 years old) is much lower than that of the northern hemisphere common minke whale Balaenoptera acutorostrata (Yasunaga et al., 2015).

The contamination of the Arctic by legacy PCBs well documented, and the recent discovery of unintentionally produced (UP) PCBs, such as PCB-5 and PCB-11, has also caused increasing concern (Bartlett et al., 2019). PCB-11 was detected in the surface snow collected from Lomonosovfonna, Svalbard, in 2009-2010 and 2013-2014, accounting for 4% of the total PCBs (O. Garmash et al., 2013). PCB-11 and other UP-PCBs are not well understood in the Arctic. Although their concentrations were not as high as those of some legacy PCBs, the toxicity of the parent source and metabolites is still unclear, which might cause a higher negative effect risk.

5.4 Environmental pathways

Fugacity ratios (*FR*s) can be used to determine the transfer of a chemical in different phases. According to the PCB *FR*s of air–soil, air-plant, air-bird guano and air-reindeer feces, the net transport direction of PCBs in Ny-Ålesund was from air to soil, while guano and feces released some PCBs into the air (Zhang et al., 2014). However, the opposite transport direction was found in the Arkona Sea. For all congeners, with the exception of PCB 180, the fugacity calculations indicated net volatilization from the surface water. This tendency was most pronounced for the lower chlorinated congeners 28/31. PCB 180, the most chlorinated congener studied, showed no consistent tendency for either volatilization or deposition. For this chemical, there appeared to be a near partitioning equilibrium between the surface water and the lower atmosphere (Bruhn et al., 2003).

The concentrations of PCBs in biota, especially in seabirds and mammals, were approximately 3 to 4 orders of magnitude higher than those in environmental media, indicating that PCBs accumulated and magnified in organisms. The ratios of bioaccumulation factors (BAF_{arctic}): BAF_{temperature}) were above 1 for all four PCB congeners in zooplankton (6.4 - 13.8) and planktivorous fish (2.9 - 5.0), whereas the ratios were below 1 in seal (Sobek et al., 2010). Both the abovementioned model and the investigation showed that temperature played an important role in PCB bioaccumulation through the food web (Cabrerizo et al., 2018; Nfon and Cousins 2007; Sobek et al., 2010). The trophic magnification factors (TMFs) were significantly larger than 1 for most PCBs and OCPs in an ecosystem on King George Island, Antarctic. The legacy POPs of PCBs, HCHs, CHLs, and DDTs were significantly magnified as the trophic level increased (TMF>1) (Kim et al., 2021).

PCBs have neurotoxicity and endocrine disruption effects in birds and mammals in polar areas. Specifically, chiral PCB congeners, in particular congeners with a 2,3,6 substitution pattern in one phenyl ring, have been linked to neurodevelopmental toxicity in humans and laboratory animals (Wu et al., 2014). Moreover, it is acknowledged that thyroid hormone (TH) synthesis is interrupted by PCBs through receptor binding and inhibits intracellular messaging after thyroid stimulating hormone (TSH) stimulation (Nøst et al., 2012). In addition, OH-PCBs have several modes of action and, e.g., can disrupt cellular calcium homeostasis by mechanisms involving RyRs (Yassaman et al., 2013) or cause thyroid dysfunction (Kodavanti and Curras-Collazo 2010; Meerts et al., 2002; Wu et al., 2014). PCB concentrations in the plasma of Canada, Russia, Norway, Greenland and the Nunavik people have been investigated for decades because PCBs are modified through the food web. Due to the similar diet of marine mammals, people living in the circumpolar region always present a high level of PCBs in plasma; in particular, the PCB levels in the blood of people in East Greenland exceed the Canadian Guideline (Deutch et al., 2007). Total PCB concentrations in the blood of pregnant Nunavik women decreased by 84% between 1992 and 2017. Nevertheless, 10% of women in 2017 had values that exceeded the guidance values. Despite the significant decline in PCBs, exposure to these contaminants is still quite prevalent among pregnant Nunavik women (Adamou et al., 2020).

6 Chlorinated paraffins (CPs)

6.1 Introduction

Chlorinated paraffins (CPs) have been widely used as flame retardants and plasticizers in industrial products and household. CPs can be divided into short-chain chlorinated paraffins (C-10-C-13, SCCPs), medium-chain chlorinated paraffins (C-14-C-17, MCCPs) and long-chain chlorinated paraffins (C->=18, LCCPs) according to the length of the carbon chain (Meziere et al., 2020). With their extensive use in people's lives, the environmental pollution caused by CPs is becoming increasingly serious. As remote areas, the level of locally sourced pollution in Antarctic and the Arctic is low, but it will be greatly affected by the atmospheric transport of POPs.

6.2 Sources and long-range environmental transport

Compared with MCCPs and LCCPs, SCCPs have lighter masses and are easier to transport through the atmosphere for a long distance to the remote Antarctic and Arctic regions, which have few anthropogenic sources. This has also been proven by previous long-term atmospheric monitoring studies (Jiang et al., 2021; Ma et al., 2014). This indicates that the pollution problem of SCCPs is becoming increasingly serious, while the pollution problem of MCCPs is relatively mild for now. At present, the relevant research reports on CPs focus on SCCPs and pay less attention to MCCPs and LCCPs, which is most likely related to their occurrence and environmental impact. SCCPs remained as a Stockholm Convention candidate for decades before they officially joined the new POP list, which shows that there has been some debate about their persistence. However, increasing evidence, especially regarding the occurrence and detection in the Antarctic and Arctic, shows that SCCPs have the general characteristics of POPs, including their potential for long-distance transport. Therefore, the pollution problem of SCCPs has become a global issue.

Production, storage, transportation, industrial application and consumer use, as well as the final disposal process of products, may cause the release of SCCPs to the environment. It is generally believed that pollutants with a half-life of more than 2 days in the atmosphere may carry out long-distance transmission. SCCPs can be adsorbed on the atmospheric particulate phase. At high latitudes and low temperatures, the oxidation pathway of SCCPs in the atmosphere is limited, which is more conducive to the long-distance transmission of SCCPs with particles. Wania (2003) predicted the Arctic pollution potential (ACP) of many SCCP congeners according to their K_{oa} and K_{aw} values. Compared with the ACP results of a series of model compounds, the ACP of SCCPs was similar to that of four to seven substituted PCBs. In addition to the long-distance transmission model data, the field experimental results supported the conclusion that SCCPs could migrate to remote areas through long-distance transport.

6.3 Occurrences in environmental matrices

6.3.1 CPs in air

Due to the high volatility of SCCPs, they were detected in the air from industrialized rgions to the polar environment. So far, CPs in the atmosphere have been investigated at several Arctic stations, e.g. Alert, Zeppelin, Barrow, Stórhöfði and Little Fox Lake (Vorkamp et al., 2019). Since 2013, CPs have been include in the monitoring program of organic contaminants at Zeppelin. The concentrations of SCCPs were usually 1-3 orders of magnitude higher than those of MCCPs (Figure 8). However, the concentration of SCCPs and MCCPs at Zeppelin in 2020 were 510 pg/m³ (range: <120-6600 pg/m³) and 750 pg/m³ (range <320-5200 pg/m³), respectively (Bohlin-

Nizzetto et al., 2021). The annual means of SCCPs were comparable from 2013 to 2020, while MCCPs showed increasing trends. In the Antarctic, the average concentration of SCCPs in air has increased by nearly two orders of magnitude in recent years, from 15 to 1200 pg/m³, while the average concentration of MCCPs has changed little, from 4.5 to 5.1 pg/m³ (Figure 9 and Table 11) (Jiang et al., 2021; Ma et al., 2014). Ma et al. collected atmospheric gas phase and particle phase samples on King George Island, Antarctic, and systematically studied the content level, homologous distribution characteristics, pollution sources and gas/solid distribution behavior of CPs in the Antarctic atmosphere. SCCPs were measured in all atmospheric samples in Antarctic, especially in the atmospheric particle and gas phases at King George Island, Antarctic. The concentration of SCCPs ranged from 9.6 to 21 pg/m³, with an average of 15 pg/m³.

Through two regression models (the Junge–Pankow model and the K_{OA}-based model), Ma et al. also found that the absorption mode of organic matter by aerosols played an important role in the distribution and migration of CPs in the atmosphere in remote areas of Antarctic (Ma et al., 2014). Lu et al. further established three different models (the J–P model, H–B model, and L–M–Y model) to estimate and measure the gas/particle distribution process of local CPs. It was found that the steady-state model (J–P model) rather than the equilibrium state models (J–P model and H–B model) better represented the gas/particle distribution process of CPs in Antarctic, which provides some help for the long-term study of CP concentration levels (Jiang et al., 2021).

Location	Sampling Time	Sample Number	Range	Mean
King George Island, Antarctic	2014-2018	120	70-4200	1200
King George Island, Antarctic	2013	24	9.6-21	15

Table 11:	Occurrence of SCCPs in air (pg/m ³) in the Antarctic
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Source: Jiang et al. 2021, Ma et al. 2014.

6.3.2 SCCPs in soil

As an environmental matrix, soil is the basis of terrestrial ecosystems. Environmental pollutants can enter and accumulate in high trophic organisms through the chain of soil-plant-herbivore-carnivore, and this is especially true for persistent organic pollutants that have lipophilic and biomagnification characteristics, such as SCCPs. The source of SCCPs in the soil in Antarctic and the Arctic is not a direct emission source; rather, it is mainly the material exchange between the soil and atmosphere. The average concentrations of SCCPs in Antarctic and Arctic soils were 14.8 and 7.1 ng/g, respectively, which proved the extensive occurrence of SCCPs in polar soils (Table 12) (Li et al., 2016, Li et al., 2017).

Table 12:	Concentrations of SCCPs (ng/g) in soil in the Arctic and the Antarctic
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Location	Sampling Time	Sample Number	Mean Concentration
King George Island and Ardley Island, Antarctic	2012-2013	8	15
Ny-Ålesund, Arctic	2011-2012	7	7.1

Source: Li et al. 2016, Li et al. 2017.





Source: Bohlin-Nizzetto et al. 2014, 2015, 2016, 2017, 2018, 2019, 2020, 2021.





Source: Jiang et al. 2021, Ma et al. 2014.

Location	Sampling Time	Tissue	Range	Mean	Note
King George and Ardley Island	2012-2013	Archeogastropoda		120	dw
King George and Ardley Island	2012-2013	Neogastropoda		89	dw
King George and Ardley Island	2012-2013	Fish		70	dw
King George and Ardley Island	2012-2013	Algae		61	dw
King George and Ardley Island	2012-2013	Moss		42	dw
Southern Ocean	2007-2015	Whales Blubber	<mdl-46< td=""><td></td><td>lw</td></mdl-46<>		lw
Hudson Bay Canada	2013-2014	polar bears liver	120-250	170	lw
Greenland	2012	Black guillemot eggs	490-940	710	ww
Greenland	2012	Glaucous gull liver	170-390	230	ww
Greenland	2014	Glaucous gull liver	170-270	220	ww
East Greenland	2012	Ringed seal blubber	910-1900	1400	ww
Greenland	2014	Ringed seal blubber	710-1200	940	ww
West Greenland	2014	Ringed seal blubber	560-1400	880	ww
Greenland	2012	Polar bear tissue	1200-2700	2200	ww
Greenland	2014	Polar bear tissue	370-1600	880	ww
Ny-Ålesund, Arctic	2011-2012	Brown algae		81	dw
Ny-Ålesund, Arctic	2011-2012	Gammarids		520	dw
Ny-Ålesund, Arctic	2011-2012	Cod		210	dw
Ny-Ålesund, Arctic	2011-2012	Moss		210	dw
Ny-Ålesund, Arctic	2011-2012	Sooty sedge		330	dw
Ny-Ålesund, Arctic	2011-2012	Arctic bell-heather		180	dw
Ny-Ålesund, Arctic	2011-2012	Mountain avens		140	dw
Sklinna and Røst island	2012-2012	Bird egg	<mdl-4.8< td=""><td></td><td>ww</td></mdl-4.8<>		ww
waters around Iceland	2001-2003	Greenland sharks liver	<mdl-5200< td=""><td></td><td>lw</td></mdl-5200<>		lw
Bear Island and Iceland	2001-2004	Liver of fish and seabird	5-88		ww
Revingehed, SkAne	1986	Rabbit muscle		2900	lw
Grimso, Vastmanland	1985-1986	Moose muscle		4400	lw
Ottsjo, Jamtland	1986	Reindeer suet		140	lw
Lake Storvindeln, Lapland	1986	Whitefish muscle		1000	lw
Kongsfjorden, Svalbard	1981	Ringed seal blubber		130	lw

 Table 13:
 Concentrations of SCCPs (ng/g) in organism in the Arctic and the Antarctic

MDL: Method detection limit; dw: dry weight; lw: lipid weight; ww: wet weight

Source: Li et al. 2016, Casa et al. 2019, Letcher et al. 2018, Vorkamp et al. 2017, Li et al. 2017, Huber et al. 2015, Strid et al. 2013, Reth et al. 2006, Jansson et al. 1993.

6.3.3 SCCPs in biota

Biological absorption of SCCPs in the environment and continuous enrichment of these pollutants in the body lead to the detection of high concentrations of SCCPs. (Casa et al., 2019; Huber et al., 2015; Letcher et al., 2018; Strid et al., 2013; Vorkamp et al., 2017). The SCCP content in environmental samples from King George Island and Ardley Island in Antarctic ranged from 3.5 to 257 ng/g, and the average concentration was 76.6 ng/g (Table 13) (Li et al., 2016). The concentration of SCCPs in the ecosystem of the Svalbard Islands in the Arctic ranged from 6.0 to 611 ng/g dw, with an average value of 165 ng/g. Among them, the concentration level of SCCPs in aquatic species was slightly higher than that in terrestrial plant samples (Li et al., 2017). The content level of SCCPs in polar regions was relatively low. Li et al. further studied the longdistance transport mechanism of SCCPs through the composition characteristics of SCCPs in the Antarctic and Arctic. The chlorinated congeners in the Antarctic and Arctic samples were mainly Cl₆ congeners, accounting for 38.0% and 34.8% of the total content of SCCPs, respectively. In terms of carbon chain length, short carbon chain (< C₁₀) congeners accounted for the main components in Antarctic and Arctic samples, accounting for 56.1% and 48.6% of the total SCCPs, respectively (Li et al., 2016; Li et al., 2017), which was consistent with the pollution characteristics of SCCPs in Arctic marine mammals (Tomy et al., 1999). Reth et al. also found that C_{10} congeners of SCCPs detected in biota in the North Sea were more abundant than those in the Baltic Sea. (Reth et al., 2006)

6.3.4 Environmental pathways

The fractionation of short carbon chain congeners occurred in the environmental samples of the North and South Poles compared with the environmental samples of middle and low latitudes and industrial products. This difference is mainly because short carbon chain congeners have higher volatility than do long carbon chain congeners, and they are more likely to travel over long distances with the atmosphere. C₁₀ congeners have a lower K_{ow} and a higher water solubility than that of other long carbon chain congeners, and they do easily adsorb and settle on water particles, so it is easier for them to migrate globally with the ocean current. The enrichment of short carbon chain and low chlorinated congeners in polar and polar environmental samples is the specific embodiment of the physicochemical properties of different congeners of SCCPs (Tomy et al., 2000).

7 Polychlorinated naphthalenes (PCNs)

7.1 Introduction

Polychlorinated naphthalene (PCN) is a series of substances composed of chlorine atoms instead of hydrogen atoms on a naphthalene ring, with 75 homologs. Chlorinated substances and polycyclic aromatic hydrocarbons all have high stability, and PCN, as a complex of them, thus has high stability and is difficult to degrade in the environment. Given its stability, PCN is widely used in various industrial applications, such as wood preservatives, paint and oil additives, cable insulation and capacitors. Because of its high toxicity, persistence, and wide detection in the environment, the presence of PCNs has been detected even in polar regions, and their biomagnification in the food chain has resulted in them being included on the new list of POPs by the Stockholm Convention (Rotander et al., 2012b). With the gradual implementation of the Convention, the production of PCNs is gradually prohibited. Although the production of PCNs as a chemical product has stopped, PCNs still exist in the industrial process of other products as a byproduct of chemical production.

7.2 Sources and long-range environmental transport

In the combustion process, polychlorinated naphthalene can be unintentionally generated in other industrial processes through a mechanism similar to PCDDs/PCDFs, such as smelting in the secondary nonferrous metal industry, cement and magnesium oxide production, aluminum refining and coking (Baek et al., 2008). In winter, when the air mass originating from Eurasia affects the high Arctic and coincides with the haze period, the PCN level is higher. Sporadic trans-Pacific transport affects the PCN concentration in Tagish. As indicated by the existence of combustion marker homologs, it is obvious that PCNs from spontaneous combustion sources contribute to the level in winter, but evaporative emissions from the source area may be the main source (Helm et al., 2004).

Location	Sampling Time	Sample Number	Range	Mean
Victoria Land, Antarctic	2010-2011	22	nd-1.6	
Alert, Canada	1994-1995	14		0.69
Tagish, Canada	1994-1995	14		0.38
Dunai, Russia	1994-1995	11		0.82
Ny-Ålesund	2001	6	27-48	35
Storsteinen Mountain	2003	10	8.7-47	25
Barents Sea	1996	5		40
Eastern Arctic Ocean	1996	25		12
Norwegian Sea	1996	4		7.1
Alert, Canada	1993-1994	7		3.5
Dunai Island, Russia	1993	4		0.84

Table 14:Occurrence of PCNs in air (pg/m³) in the Arctic and the Antarctic

Source: Barbaro et al. 2016, Helm et al. 2004, Herbert et al. 2005, Harner et al. 1998.

7.3 PCNs in environmental matrices

7.3.1 PCNs in air

Previous polar research on PCNs paid more attention to the Arctic environment, and now, more attention is being paid to the Antarctic environment. In previous Arctic air monitoring projects, PCNs were detected in the air of the Arctic regions of Canada, Russia and Norway, and high PCN contents were detected in some regions. For example, the concentrations of PCNs ranged from 27.0-48.0 pg/m³ in Ny-Ålesund and 8.70-47.0 pg/m³ in Tromso (Herbert et al., 2005), and the average concentration was 40.4 pg/m³ in the Barents Sea (Harner et al., 1998) (Figure 10 and Table 14). In addition, PCNs were detected in the snow in these areas. These results indicate that the occurrence of PCNs in the Arctic is relatively common, and the higher concentration in some areas indicates that PCNs may be affected by local source emissions from nearby towns.

Figure 10: Concentrations of PCNs (mean, pg/m³) in air samples (gaseous and particle phases) in the Arctic and Antarctic



Source: Harner et al. 1998, Helm et al. 2004, Herbert et al. 2005, Barbaro et al. 2016.

In the research project of remote Arctic monitoring stations in Alert and Tagish, Canada, and Dunai Island, Russia, the annual average concentrations of PCNs of Alert, Dunai and Tagish were 0.69, 0.82 and 0.38 pg/m³, respectively. Meanwhile, PCNs showed a seasonal trend in Alert and Dunai. In winter, when the air mass originating from Eurasia affects the high Arctic and coincides with the haze period, the PCN level is higher. Sporadic trans-Pacific transport affects the PCN concentration in Tagish. As indicated by the existence of combustion marker homologs, it is obvious that PCNs from spontaneous combustion sources contribute to the level in winter, but evaporative emissions from the source area may be the main source (Helm et al., 2004).

However, in the Antarctic, the detected PCN concentration was very low. In the aerosol samples collected at a coastal site (Terra Nova Bay, Victoria Land) in Antarctic, only nd-0.6 pg/m³ PCN was detected (Figue 10) (Barbaro et al., 2016).

Location	Year	Tissue	Range	Mean	Note
Alaska Arctic	1997-1999	Polar bear liver		0.37	ww
Ross Sea	1994-1999	South polar skua liver		2.6	ww
Ross Sea	1994	Krill		0.0015	ww
Ross Sea	1995	Sharp-spined notothen	0.0013-0.0028		ww
Ross Sea	1995	Icefish	0.0021-0.0047		ww
Ross Sea	1994	Silverfish		0.086	ww
Ross Sea	1996	Weddell seal liver		0.077	ww
Ross Sea	1997	Weddell seal blubber		0.044	ww
King George Island	2013-2014	Limpet	0.024–0.20		lw
King George Island	2013-2014	Antarctic cod	0.23–0.53		lw
King George Island	2013-2014	Amphipod	0.22–0.60		lw
King George Island	2013-2014	Antarctic Icefish		1.8	lw
King George Island	2013-2014	Gentoo penguin	0.012–26		lw
King George Island	2013-2014	Chinstrap penguin	0.16–1.5		lw
King George Island	2013-2014	South polar skua		58	lw
King George Island	2013-2014	Kelp gull		2	lw
Terra Nova Bay	1996-2009	Scallop tissue	0.004-0.63		dw
King George Island	2008-2009	Penguin	0.69–2.1		lw
King George Island	2008-2009	Skua	7.4–180		lw
Southern Hudson Bay	2013–2014	Polar bear liver and fat	12-57	27	ww
Western Hudson Bay	2013–2014	Polar bear liver and fat	8.8-46	20	ww
Prince Leopold Island	1975-2014	Khick-billed murres egg	0.36-1.0		ww
Livingston Island	2004	Antarctic fur seal blubber		1.3	ww
Northern Gulf of Alaska	2000-2001	Harbor seal	0.3-27		lw

Table 15:Occurrence of PCNs (ng/g) in organism in the Arctic and the Antarctic

Source: Braune 2017, Corsolini et al. 2002, Helm et al. 2002, Kim et al. 2015, Kim et al. 2021, Letcher et al. 2018, Schiavone et al. 2009a, Wang et al. 2007.

7.3.2 PCNs in biota

In the Ross Sea in the Antarctic, researchers collected a variety of biological samples of krill, spiny vertebrates, crocodile ice fish, Antarctic silverfish, Antarctic skunk and Weddell seal and detected the occurrence of PCNs. Although the concentration was very low, e.g., only 1.5 pg/g ww PCNs was detected in krill biomass, this was the first study to record the occurrence of PCNs in Antarctic organisms. This result indicates that PCNs have reached the Antarctic continent without local emission sources in various ways (Corsolini et al., 2002). In penguin samples obtained on King George Island in Antarctic, the highest detected concentration of PCNs increased in recent years, and their concentration range has increased from 0.69–2.07 ng/g lw to 0.012-26 ng/g lw (Kim et al., 2021; Kim et al., 2015). Grotti et al. analyzed 80 scallop tissue samples collected and stored in the Antarctic environmental sample bank from 1996 to 2009 through retrospective monitoring. The concentrations of PCNs were consistent with other literature reports, and no temporal trend was found, indicating that the pollution level of these compounds was low (Grotti et al., 2016). In a long-span study of the Arctic, Braune et al. determined the concentration of PCNs in thick-billed crow eggs collected from Prince Leopold Island in the Canadian Arctic between 1975 and 2014. From 1975 to 2014. The concentration of PCNs in sea fish eggs decreased significantly at an average annual rate of 15 pg/g ww (Braune and Muir 2017).

The PCNs in organisms in the Arctic and Antarctic are summarized in Table 15. High concentrations of PCNs have also been detected in marine mammals in the Arctic, such as in polar bears, seals and whales (Helm et al., 2002; Letcher et al., 2018; Wang et al., 2007). Helm et al. measured the PCNs in marine mammals in the Canadian Arctic for the first time. The concentrations of PCNs in beluga whale blubber were 36-383 pg/g lw and those in ringed seal were 16-71 pg/g lw (Helm et al., 2002). Wang et al. first reported PCN contamination in Alaskan ringed seal tissues. From 2000 to 2001, a variety of biological tissues, including fat, liver and kidney samples of ringed seals in the northern Gulf of Alaska, were collected. The total detected PCN concentration ranged from 0.3 to 27 ng/g lw. It was also found that the PCN concentration was related to the age, sex, weight and fat thickness of ringed seals (Wang et al., 2007). In 2013-2014, Letcher et al. collected polar bear fat or liver tissue samples in southern and western Hudson Bay and investigated 295 residual and newly halogenated persistent organic pollutants (POPs). The PCN concentration detected in West Hudson Bay was 8.8-46 ng/g ww and 12-57 ng/g ww in South Hudson Bay (Letcher et al., 2018).

8 Dioxins

8.1 Introduction

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are toxic compounds from many kinds of sources and persistent in the environment. They are very stable, resistant to chemical and biological decomposition mainly bind to solid substrates such as soil and sediments. PCDD and PCDF have similar physicochemical properties. For PCDD, two oxygen bridges connect the benzene ring, while in the case of PCDF, one carbon bond and one oxygen bridge connect the benzene ring. They contain four to eight chlorine atoms.

8.2 Sources and long-range environmental transport

PCDDs and PCDFs are produced by incomplete combustion and unintentionally in the manufacture of pesticides and other chlorinated substances. They mainly come from the combustion of hospital waste, urban waste and hazardous waste, as well as from automobile exhaust, peat, coal and wood (Zhang et al., 2017). PCDDs/PCDFs can accumulate in organisms through the food chain (Schoeters and Hoogenboom 2006). In addition, the atmosphere has become the most important way for PCDDs/PCDFs to be transported from emission sources to remote environments (Kampa and Castanas, 2008).

8.3 Dioxins in environmental matrices

8.3.1 Dioxins in seawater

The pollutant distribution of suspended particulate matter samples in Arctic fjord seawater was investigated. A total of 17 kinds of PCDDs/PCDFs were found, with a total concentration range of 0.066-230 pg/L, of which highly chlorinated PCDFs constituted the most important part. The high concentration of dioxins detected in some samples showed that these pollutants might pose a potential threat to the marine biological community (Pouch et al., 2021).

Location	Sampling Time	Sample	Sample Number	Range	Mean
Ardley Island	2010	Soil	36		11
Ardley Island	2010	Ornithogenic soil	18		11
Fildes Peninsula	2007-2008	Soil	15	0.49-6.7	2.2
NyÅlesund	2008-2008	Soil	20	3.6–17	10

Table 16:Occurrence of Total PCDD/Fs (pg/g, dw) in soil from the Arctic and the Antarctic

Source: Mwangi et al. 2016, Jia et al. 2014.

8.3.2 Dioxins in soil

Through a comparative study, it was also found that the concentration of PCDD/PCDF in soil samples near the research bases was higher than that of PCBs in the vicinity of Zhongshan Station in Antarctic, China. These phenomena may be caused by combustion related to research activities, such as waste and fuel combustion for power generation and household heating (Mwangi et al., 2016). In a comparative study of Antarctic, the Arctic and the Tibetan Plateau, 35

soil samples collected from the Fildes Peninsula in the Antarctic and from Ny-Ålesund in the Arctic were analyzed. Among them, the total average concentration of PCDD/PCDF was 10 pg/g in Ny-Ålesund and 2.2 pg/g in Fildes Peninsula, indicating that human activities played a leading role in the emission of this kind of pollutant (Table 16). The distribution of PCDD/PCDF homologs and homologs in the polar regions was dominated by highly chlorinated PCDD/PCDF because they emit more and remove more slowly than less chlorinated homologs (Jia et al., 2014).

8.3.3 Dioxins in biota

Because of their strong toxicity, the detection of trace amounts represents a great risk to the ecological environment. As remote areas, the Arctic and Antarctic are less affected by man-made combustion sources. However, the presence of PCDDs/PCDFs has been detected in these areas, indicating that these areas have been affected by the long-distance atmospheric transport of these persistent organic pollutants. As the largest marine reserve in the world, the Ross Sea has a complete ecosystem. Through sampling and analysis of a variety of species in the ecosystem, PCDDs were found in many samples, and PCDFs were detected only in biological samples of some species. The average concentrations of PCDDs and PCDFs detected in the muscle of Antarctic toothfish were 10 ng/g ww and 12 ng/g ww, respectively (Corsolini et al., 2017). Krill plays an important role in the basic food web of the Southern Ocean. It transfers persistent organic pollutants in phytoplankton to the animal food chain. The impact of these pollutants on the whole animal food web can be effectively evaluated through the investigation of pollutants in krill. In the detection of pollutant levels in krill in the Bellingshausen, South Skosher and Weddell Seas around the Antarctic Peninsula, the total concentration range of PCDD/PCDF was 6.2 pg/g lw to 160 pg/g lw (Galban-Malagon et al., 2018). In the vicinity of Zhongshan Station in Antarctic, China, dead penguins were accidentally found during the expedition, and the occurrence of PCDD/PCDF was also detected in the penguins. The total concentration found in Adelie penguins was 430 pg/g lw. In addition, 55, 28, 22, 19 and 40 pg/g lw were detected in the brisket, back leg fat, dominant fat, breast fat and river of emperor penguin, respectively (Table 17). The concentration detected in the lichen samples was only 1.8 pg/g lw (Mwangi et al., 2016). The detection of PCDD/PCDF in Antarctic penguins shows that such pollutants have entered the body of Antarctic animals universally (Corsolini et al., 2007), but the detection in penguin eggs shows that penguin offspring have a certain concentration of pollutants from the beginning of hatching. Adelie penguins were more vulnerable to the accumulation of pollutants in the environment than were gentoo and chinstrap penguins, and only a small amount of PCDD/PCDF was found in their body, but it may cause serious problems in some aspects (Schiavone et al., 2009a).

Arctic cod liver, as a traditional food in Norway, is a potential source of POPs in the diet of local residents. Through the sampling and analysis of wild cod in the Arctic, it was found that the total average concentration of PCDD/PCDF was 14 pg/g ww. The extremely low pollutant concentration level indicates that its potential risk is very low (Julshamn et al., 2013). High trophic animals are generally prone to accumulate high concentrations of persistent organic pollutants in vivo due to biomagnification. As a high trophic fish in Arctic waters, the content of PCDD/PCDF in Greenland shark also reached a high concentration. In the biological samples of Greenland shark collected in Icelandic waters from 2001 to 2003, the average concentration in muscle was 13 pg/g lw and that in liver was 530 pg/g lw. In addition, an enrichment effect was found in the liver tissue (Strid et al., 2007).

Location	Year	Sample	PCDDs	PCDFs	PCDD/Fs	Note
Antarctic Peninsula	2009	Krill			54	lw
North-East Atlantic	2001-2003	Shark liver	71	460	530	lw
North-East Atlantic	2001-2003	Shark muscle	2.9	10	13	lw
Livingston Island	2004-2004	Fur seal blubber	54	96		ww
Livingston Island	2004-2004	Fur seal liver	11	150		ww
Livingston Island	2004-2004	Fur seal muscle	3.5	85		ww
Alaska	1993-2000	polar bear liver			26	lw
Edmonson Point	1995-1996	Penguin egg			23	lw
Terra Nova Bay	1998-1999	South polar skua egg			180	Iw
Edmonson Point	1995-1996	Weddell seal liver			8.9	lw
Terra Nova Bay	1994-1995	Krill			27	lw
Terra Nova Bay	1994-1995	fish				lw
Larsemann Hills	2009	Adelie penguin			430	Iw
Larsemann Hills	2009	Penguin Brisket			55	lw
Larsemann Hills	2009	Penguin Back leg fat			28	Iw
Larsemann Hills	2009	Penguin Abdominal fat			22	lw
Larsemann Hills	2009	Penguin Breast fat			19	lw
Larsemann Hills	2009	Penguin Liver			40	lw
Larsemann Hills	2009	lichen			1.8	lw
U.S. Lenie Field Station	2004-2004	Adelie penguin blood	22	50		ww
U.S. Lenie Field Station	2004-2004	Chinstrap penguin blood	6.5	2.5		ww
U.S. Lenie Field Station	2004-2004	Gentoo penguin blood	18	9.2		ww
Terra Nova Bay	2002	Antarctic toothfish muscle	10	12		ww
Barents Sea	2009-2010	Arctic cod liver	1.1	1.4	2.4	ww
Barents Sea	2009-2010	Arctic cod muscle			0.045	ww
Barents Sea	2012-2012	Red king crab	0.08	0.02	0.04	ww

Table 17:Concentrations of Dioxins (pg/g) in organism in the Arctic and the Antarctic

*=TEQ (Toxic Equivalent Quantity, WHO-2005) concentration

Source: Galban-Malagon et al. 2018, Strid et al. 2007, Schiavone et al. 2009a, Kumar et al. 2002, Mwangi et al. 2016, Corsolini et al. 2007, Corsolini et al. 2017, Julshamn et al. 2013, Julshamn et al. 2015.

9 Per- and polyfluoroalkyl substances (PFASs)

9.1 Introduction

Poly- and perfluoroalkyl substances (PFASs) are a class of chemicals composed of fluorinated aliphatic carbon skeletons and hydrophilic groups, which endow PFASs with unique properties, such as both hydrophobic and oleophobic properties and excellent physical and chemical stability. Thus, PFASs are extensively used in fire-fighting foams, surfactants and metal-plating industries ((Buck et al., 2021). PFOS and PFOA are the most widely known PFAS because of their presence in the global environment, from source regions to remote areas (Dietz et al., 2008; Giesy and Kannan 2001; Shoeib et al., 2006). Laboratory studies have proven that PFAS exposure may cause chronic health effects and toxicities (Balan et al., 2021; Hoover et al., 2019; Lau et al., 2007; Podder et al., 2021). The annual global production of PFASs was estimated to be 4400-8000 tons/year between 1975 and 2004 (Prevedouros et al., 2006). PFASs may directly enter the environment during the production, application of firefighting foams, and emission from commercial products (Schultz et al., 2003). The major PFASs determined in the polar regions are listed in Table A15.

9.2 Source and long-range environmental transport

Due to their unique properties, PFASs can be transported from sources to remote areas through both atmospheric and ocean currents and have been widely present in all environmental compartments in the remote Arctic and Antarctic (Muir and Miaz 2021; Xie et al., 2015). Neutral PFASs, such as FTOHs, Me/EtFOSE and FOSE, mainly partition into air and travel with air masses from source regions to remote areas (Lin et al., 2020a; Taniyasu et al., 2013; Wang et al., 2015c). 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). In the polar regions, photodegradation and reaction with OH, Cl or O₃ radicals are the major pathways to eliminate neutral PFASs from the atmosphere (Ellis et al., 2004a; Ellis et al., 2003; Loewen et al., 2005), especially in spring and summer. The accumulation of PFCAs and their precursors in Arctic snow and ice has been observed in Ny-Ålesund and the high Arctic (Garnett et al., 2021; Xie et al., 2015). The presence of perfluorodecanoic acid (PFDA) and perfluoroundecanoic acid (PFUA) on the ice cap indicated atmospheric oxidation of the precursors is an important source (Young et al., 2007).

Neutral PFASs such as perfluorosulfonamides (FOSA), perfluorosulfonamidoethanols (FOSE) and FTOHs are the precursors of ionic PFASs (D'Eon et al., 2006; Ellis et al., 2004b). Because of the low reaction rates with hydroxyl radicals, the atmospheric lifetimes of FTOHs and FASAs ranged 10–50 days (Ellis et al., 2004b; Martin et al., 2006a; Piekarz et al., 2007), which allow they undergo LRAT, e.g., to the Arctic and Antarctic (Dreyer et al., 2009; Stock et al., 2007). The neutral precursors can be a important source of PFOA and PFOS in remote regions (Andersen et al., 2005; Ellis et al., 2004b; Martin et al., 2006a). Besides, FTOHs and FTSEs/FOSAs in the aquatic phase can transform to PFCAs and PFSAs (Armitage et al., 2006, 2009, Cousins et al., 2011). In addition, increasing human activities in the Arctic and Antarctic may directly release PFASs into polar environments.

9.3 Occurrences in environmental matrices

9.3.1 PFASs in air

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). Atmospheric transport is the primary pathway by which neutral PFASs migrate to polar regions from source areas (Prevedouros et al., 2006; Wang et al., 2015c). Neutral PFASs determined in the environmental samples include 6:2, 8:2, 10:2 FTOHs, 6:2, 8:2, 10:2 fluorotelomer acrylates (FTAs), methyl and ethyl perfluoro-octane sulfonamides (MeFOSA and EtFOSA), and methyl and ethyl perfluoroctane sulfonamidoethanols (MeFOSE and EtFOSE) (Cai et al., 2012b; Dreyer et al., 2009; Wang et al., 2015c).

The neutral PFASs in the Southern Ocean and along the Antarctic Peninsula ranged from 3.7 to 48.1 pg/m³ (Del Vento et al., 2012; Wang et al., 2015c). Among FTOHs, 8:2 FTOH (mean: 12.7 \pm 9.8 pg/m³) was the most predominant, followed by 10:2 FTOH (4.2 \pm 3.3 pg/m³), 12:2 FTOH (1.8 \pm 1.2 pg/m³), and 6:2 FTOH (0.5 \pm 0.5 pg/m³) (Figure 11). Two fluorotelomer acrylates, 8:2 FTAC and 6:2 FTAC were determined in the Southern Ocean, with concentrations ranging from <0.01–0.08 pg/m³ and <0.04–0.25 pg/m³, respectively. In the Antarctic Peninsula, 6:2 FTAC was determined in three air samples (0.37–1.8 pg/m³).

Figure 11: Neutral PFASs are present in air from the Southern Ocean and Antarctic Peninsula. FTOHs are dominant components among the neutral PFASs, followed by N-MeFBSA and N-MeFOSE.



Source: Del Vento et al. 2012, Wang et al. 2015c.

Studies on neutral PFASs in air have been carried out in the Arctic based on stationary monitoring programs and expedition cruises in the Arctic and adjacent oceans. In the Canadian Arctic (Shoeib et al., 2006), the sum concentrations of gas- and particle-phase FTOHs were 8:2 FTOH (5.8-26 pg/m³), followed by 10:2 FTOH (1.9-17 pg/m³) and 6:2 FTOH (BDL to 6.0 pg/m³). Interestingly, relatively high concentrations of EtFOSE (2.6 to 31 pg/m³) and MeFOSEA (BDL to 8.9 pg/m³) were also present in the atmosphere in the Canadian Arctic. Cai et al. reported

elevated neutral PFAS levels in oceanic air from the North Pacific to the high Arctic onboard Chinese ice breaker *Snow Dragon I* from July to August 2010 (Cai et al., 2012b).

Neutral PFASs have also been determined in air samples collected at Arctic stations at Alert, Villum, and Ny-alesund (Muir et al., 2019; Bossi et al., 2016). At the Villum Station, the average sum of the seven neutral PFASs (\sum_7 PFAS) ranged from 1.8 to 32 pg/m³. A similar level of neutral PFASs was reported in the atmosphere of Ny-Ålesund (Xie et al., 2015), where 12 neutral PFASs ranged from 6.7 to 39 pg/m³. In the Canadian Arctic (Wong et al., 2018), the concentration of FTOHs in Alert ranged from <0.17–30 pg/m³ with a median of 3.8 pg/m³. The levels of FOSEs at Alert were higher than the levels of FOSAs, with median and range concentrations of 0.49 (< 0.10–4.8) pg/m³ and 0.13 (< 0.014–0.82) pg/m³, respectively. FTOHs showed clear seasonal trends with increasing concentrations in the summer months and lower concentrations in autumn and winter in the Arctic. The seasonal trends of FOSEs, FOSA and FTAs were not remarkable because of their low levels and high variation in air.

For ionic PFASs, both PFAAs and PFSAs have been determined at very low concentrations in the atmosphere in the Arctic and Antarctic. Ionic PFASs are likely partitioned to the particle phase in the atmosphere due to their physicochemical behaviors. At the Alert research station (Wong et al., 2018), among ionic PFAS, PFBA (99%, median: 0.097 pg/m³) was the most detectable compound in air, followed by PFOS (96%, median: 1.7 pg/m³) and PFOA (89% median: 0.07 pg/m³).

Ionic PFASs have been included in Norwegian atmospheric monitoring programs at Zeppelin and Birkenes since 2006 and at Andøya since 2009 (Muir et al., 2019). The annual mean concentration of PFOA in 2020 was 0.12 pg/m³ in Birkenes, which was 2 times higher than that in Andøya (0.05 pg/m³) and Zeppelin (0.07 pg/m³) (Bohlin-Nizzetto et al., 2021). Other ionic PFAS, such as PFDA, PFNA, PFHpA, PFHxA, PFBS, PFDS and 6:2 FTS, were detected with low frequency, and the concentrations were close to the method detection limits.

The atmospheric concentrations of PFOS at Alert, Zeppelin, and Andøya were consistent, which could represent the background level of PFOS in the Arctic (Wong et al., 2018). The PFOA concentrations found at Zeppelin and Andøya were 3 times higher than those measured at Alert. Considering the distance between the sampling station and the coast, PFOA present in air could be interfered with by oceanic sources, such as marine spray and aerosols. The atmospheric concentrations of PFBA measured at Alert were comparable to those from urban sites and other remote air, suggesting that PFBA might favor long-range atmospheric transport. However, the transformation of certain volatile hydrofluorocarbons (HFCs) and hydrofluoroethers (HFEs) to PFBA might account for the elevated level of PFBA in the Arctic, which has been supported by laboratory studies (Wong et al., 2018).

In the Southern Ocean and Antarctic continent, there are limited data available regarding the amount of ionic PFASs in air. Along the Atlantic transect from Bremerhaven to Cape Town, PFOS were detected in two parallel air samples collected near South Africa, with concentrations of 0.05 and 0.24 pg/m³, which were very close to the MDL of PFASs (MDL: 0.05 pg/m³) (Jahnke et al., 2007). In the Southern Ocean, 12 PFAS were detected in air, with contents ranging from 1 to 5 pg/m³ (3 pg/m³) (Yamazaki et al., 2021). Shorter chain PFCAs, such as PFBA, PFPeA, and PFHxA, dominated in the air of the Southern Ocean, with mean concentrations of 0.80 \pm 0.52, 0.45 \pm 0.29, and 0.38 \pm 0.24 pg/m³, respectively.

9.3.2 PFASs in snow and ice

Ionic PFASs have been determined in surface snow from Longyearbyen and Ny-Ålesund in Svalbard (Kwok et al., 2013). Elevated PFAS concentrations were present in snow toward

Longyearbyen, implying the impact of local emissions. PFBA, PFOA, and PFNA were dominant in ice core samples, while PFOA was the main PFAS in surface snow. PFOA and PFNA were the main PFASs measured in surface snow samples collected from Greenland, with average concentrations of 0.53 and 0.45 ng/L, respectively.

The average concentrations of dominant PFBA light snowpacks were 2700 ± 3200 pg/L in 2013 and 2600 ± 680 pg/L in 2014, respectively. PFBA was the dominant PFAS in snow from Devon Ice, with concentrations ranging from 120 to 2000 pg/L, followed by PFNA (30-1420 pg/L), PFOA (70-680 pg/L) and PFDA (10 to 230 pg/L) (MacInnis et al., 2017). An unknown source rather than atmospheric oxidation of FTOHs might control the presence of PFBA in the snow. Moreover, ionic PFASs have been measured in snow from northern Sweden (64°N, 19°E) and northern Norway (Muir et al., 2019).

In the Antarctic, 16 PFASs have been measured in surface snow samples from Dome C on the Antarctic Plateau (Xie et al., 2020). The ΣPFASs ranged from 700 to 1800 pg/L, with a mean of 1100 ± 280 pg/L. Long-chain PFCAs (C9-C14) accounted for 10% of ΣPFCAs, which might originate from LRAT and the degradation of their volatile precursors, such as 8:2 FTOH, 10:2 FTOH and 12:2 FTOH (Wang et al., 2015c). PFOS was the predominant PFSA, with concentrations ranging from 36 to 62 pg/L, followed by PFBS (17-35 pg/L) and PFHxS (2.6-11.9 pg/L). HFPO-DA was found in Antarctic snow for the first time, ranging from 4.7 to 13 pg/L. The presence of both legacy and emerging PFASs inland Antarctic has proven that ionic PFASs can be transported to remote Antarctic and undertake environmental cycles in polar regions.

Casal et al. investigated PFASs in fresh snow, snowmelt, seawater and plankton samples collected at Livingston Island from December 2014–February 2015 (Casal et al., 2017). PFOA and PFOS were detected in 100% of fresh snow samples, and the concentrations ranged from 29-1300 pg/L and 2.1-110 pg/L, respectively. PFBA was determined in 74% of the snow samples, with concentrations ranging from below the MDL to 530 pg/L. Long-chain PFASs, such as PFNA, PFDA, PFUnDA, PFDoDA and PFTrDA, were determined in snow samples with concentrations ranging from 14-330 (68%), 3.1-600 (100%), 2.0-150 (95%), 0.8-180 (90%), and 0.74-9.8 pg/L (63%), respectively. PFTeDA was detected in only one snow sample with a concentration of 20 pg/L. The average concentration of PFBA was 420 pg/L, and contributed more than 75% to ΣPFASs in snowmelt, which might be caused by early elution of the shortest-chain PFCAs from melting surface snow.

Cai et al. reported PFASs in snow and lake waters in Fildes Peninsula, King George Island, in 2011, with total PFAS concentrations ranging from 1129 to 2491 pg/L in surface snow(Cai et al., 2012c). PFBA was the predominant PFAS (1714-2670 pg/L) in snow and lake water and accounted for more than 50% of the total PFAS, followed by PFHxA (18%) and PFOA (12%).

9.3.3 PFASs in river water and lake water, seawater

The mean concentration of Σ_{15} PFASs was 190 pg/L in ice melting lakes of Larsemann Hills, East Antarctic, and PFOA dominated with a 30% contribution and a mean of 56 pg/L (Shan et al., 2021b). The 6:2 Cl-PFESA, an emerging substitute of PFOS, was reported for the first time in the lake water of Antarctic with concentrations ranging from <11–15 pg/L.

In the Arctic, PFASs have been determined in lake water from the Canadian Arctic, Faroe Island and Svalbard (Stock et al., 2007). Lescord et al. determined PFAS concentrations in Resolute and Meretta Lakes and identified PFECHS and fluorotelomer sulfonates (FTS) (4:2-, 6:2-, and 8:2 FTS) (Lescord et al., 2015). C4-C10 PFCA was measured in four lakes on the Faroe Islands, and the concentrations were comparable with those in remote lakes (Eriksson et al., 2013).
Kwok et al. reported PFCAs in the river water at Longyearben (Kwok et al., 2013). Skaar et al. measured PFASs in lake and run-off water from firefighting training stations (FFTS) in Longyearben and Ny-alesund (Skaar et al., 2019). The concentrations of Σ_{14} PFAS ranged from 0.4 to 4 ng/L in surface lake water, 5–6 ng/L in seawater, and 110–120 ng/L in run-off water. PFOS was the most abundant compound and contributed 60–69% of Σ PFASs. Ali et al. (Ali et al., 2021a) found that PFOS (19 ± 0.87 ng/L) was the predominant PFAS in FFTS-creek, which receives runoff from FFT where AFFF is actively used. 6:2 FTS was detected with a mean of 1.5 ± 0.08 ng/L, which has been used to replace PFOS after the phase–out in the 2000s.

There are a few surveys of PFASs in oceanic seawater from the Southern Ocean and Antarctic coast. Shan et al. detected PFASs in 39% of seawater samples (n=9) in the Southern Ocean, with total concentrations of Σ_{14} PFASs ranging from 0.06-0.09 ng/L (Shan et al., 2021a), which was comparable to the PFASs measured in seawater from the Atlantic to the Southern Ocean in 2006 and 2010 (< 0.05 ng/L) (Ahrens et al., 2010; Zhao et al., 2012) and lower than those measured in the Antarctic Ocean in 2012 (PFOA: 160 ± 79 pg/L, PFBA: 100 ± 26 pg/L) (Yamazaki et al., 2021) and the coastal area of the Antarctic (mean of Σ PFAS: 190 ± 79 pg/L) (Casal et al., 2017).

In the Arctic Ocean, PFASs have been investigated in seawater along the Pacific-Arctic and Atlantic-Arctic transects. PFASs were measured in the North Pacific to the Arctic Ocean with a mean of 500 ± 170 pg/L, which was slightly lower than that in the Northwest Pacific Ocean (560 ± 170 pg/L) but obviously higher than that in the Bering Sea (340 ± 130 pg/L) (Cai et al., 2012d), and PFASs were measured in the Greenland Sea (Σ PFAS: 48 to 270 pg/L) (Zhao et al., 2012). It is estimated that 1.7–5.9 t/year of PFOA could be transported from the high Artic to the Atlantic through the Farm Strait. Joerss et al. detected HFPO-DA in seawater of the Norwegian Sea with concentrations ranging from <6.0 to 70 pg/L (Joerss et al., 2020). Among PFCAs, PFOA (49-95 pg/L), PFHxA (39-78 pg/L), PFHpA (26-58 pg/L) and PFNA (29-38 pg/L) were 100% detected in Arctic seawater, with concentrations at similar levels. PFBS was the predominant PFSA, with concentrations ranging from 23-64 pg/L, while PFOS was below the method detection limit in most seawater samples. The vertical profiles showed higher PFAS concentrations in the surface water layer than in intermediate waters and a negligible intrusion into deep waters (>1000 m).

Garnett et al. investigated PFASs in pack ice, pond water and near-surface seawater at two icecovered stations located north of the Barents Sea (81 °N)(Garnett et al., 2021). PFOA was the dominant PFCA in the near-surface seawater, with a mean concentration of 930 \pm 620 pg/L, which was higher than that recently measured in surface waters of the North Sea that received industrial discharge (Joerss et al., 2020). PFBA was the predominant short-chain PFAS and contributed more than 80%. The data of the long-chain PFASs were more complicated, with maximal concentrations measured in near-surface seawater (1600 \pm 1200 pg/L), which was 10 times higher than those reported in the Norwegian Sea (Joerss et al., 2020). The complexity of the data and composition profile in seawater, sea ice and snow requires further attention.

Generally, PFASs were present above 150 m depth in the polar mix layer (Yeung et al., 2017). PFOA and PFOS were the predominant PFASs in the vertical profiles in the Amundsen Basin and Nansen Basin, with mean concentrations of 50 and 47 pg/L, respectively. Benskin et al. reported PFHxA, PFHpA, and PFOA to be the main PFAS in Canadian Arctic Islands and Beaufort/Chukchi waters (Benskin et al., 2012). PFBA and PFHpA were the dominant PFASs in seawater at the Barrow Strait in the central Canadian Arctic Archipelago and in Adventfjorden/Isfjorden, Svalbard.

9.3.4 PFAS in sediment

Lin et al. investigated PFASs in the Bering Sea and Chukchi Sea (Lin et al., 2020b). The mean concentrations of Σ 9PFASs were 0.8 ± 0.55 ng/g in the Bering Shelf, 0.64 ± 0.45 ng/g in the Chukchi slope, 0.56 ± 0.28 ng/g in the Chukchi shelf and 0.20 ± 0.18 ng/g in the Canadian Basin. PFOS was the predominant PFAS and accounted for 35%, followed by PFNA (28%), PFBS (16%) and PFHxS (16%). PFOA and PFDA had only 2-3% contributions in the sediment. No clear temporal trends from 1975 to 2015 were observed in the sediment cores. Macinnis et al. investigated 23 PFASs in low and high Arctic lakes in Canada (MacInnis et al., 2019). Emerging PFASs such as PFECHS, ADONA, 8-Cl-PFOS, Cl-PFESAs, 6:2 Cl-PFESA and 8:2 Cl-PFESA were detected in two sediment cores collected from Lake Hazen in 2012 and Lake B35 in 2009. Increasing fluxes of PFAS in sediment cores from Lake Hazen can result from the enhanced flux of sediment and the release of historical archived PFASs in glaciers melting from the island (MacInnis et al., 2019).

PFASs have been found in FFTS surface soil with concentrations of 1140 ng/g dw near FFTS and 2.1-7.1 ng/g dw 500 m away. PFOS was the predominant PFAS with contributions > 90% (Skaar et al., 2019). Concentrations of PFASs were 81.65 ± 2.13 ng/g in landfills and 4.61 ± 3.92 ng/g in Adventfjord (Ali et al., 2021b). Furthermore, 6:2 FTS was found in sediment samples collected near the active FFTS with a mean concentration of 4.0 ng/g and a contribution of 86% of the total 14 PFAS.

Presently, few data are available for both neutral and ionic PFASs in sediment from the Antarctic and the Southern Ocean. Given the measurements for PFASs in air, seawater, ice-melt lakes and snow deposition, PFASs, especially long-chain ionic PFASs, can also accumulate in the sediment in the Antarctic, which requires further study.

9.3.5 PFAS in biota

Generally, relatively low concentrations of PFAS were present in the food web of the Antarctic (Giesy and Kannan 2001; Tao et al., 2006; Schiavone et al., 2009b). Nash et al. investigated PFASs in blood, liver and tissue samples of several species in the Antarctic region (Nash et al., 2010). PFOS and PFUnDA were the predominant PFASs in most species. Rotti et al. measured 18 PFASs in the blood plasma of adult lactating Weddell seals (Leptonychotes weddellii) from McMurdo Sound, Antarctic (Routti et al., 2015). Cocentrations of PFUnDA ranged from 0.08 to 0.23 ng/ml. This was the first study reporting detectable PFASs in a relatively nonmigratory marine mammal at high latitudes in Antarctic. Munoz et al. (Munoz et al., 2017) analyzed 26 PFASs in seabird plasma from Antarctic, including C5–C14 PFCAs, C4, C6, C7, C8, C10 PFSAs, FOSA, MeFOSA, EtFOSA, MeFOSAA, EtFOSAA, 4:2 FTS, 6:2 FTS, 8:2 FTS, polyfluoroalkyl phosphate diesters (diPAPs) and perfluoro-octane sulfonamide phosphate diester (diS-AmPAP). Eleven of the 26 PFASs were detected in seabird plasma, with total concentrations ranging from 0.37–19 ng/g. Midthaug et al. reported a PFOS concentration of 5.08 ng/g in whole blood from south polar skua (Catharacta maccormiciki) from 2013/2014 in Dronning Maud Land (Midthaug et al., 2021). Higher concentrations of PFAS were often found in the blood plasma or liver of migratory birds. However, the report of elevated levels of PFASs in south polar skua during the breeding season implied that bioaccumulation of PFASs could occur through the food chain.

Muir et al. comprehensively summarized PFASs in biota for terrestrial environments, freshwater systems, and marine environments in the Arctic (Muir et al., 2019). The distribution pattern of PFASs in vegetation and animals was diverse at the tissue and trophic levels. In general, PFOA, PFNA and PFDA predominated in vegetation, while PFNA, PFOS, PFUnDA, PFDA, PFTrDA and PFHxA were the main PFASs 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 FTSAs, were detected in the liver of Arctic foxes from Svalbard with frequencies of 33 and 13%, respectively. Bossi et al. carried out a systematic investigation of PFASs in biota from Greenland (Bossi et al., 2015). PFNA, PFDA, PFUnDA and PFDoDA were the predominant PFASs . In muskox liver samples, PFNA, PFDA, and PFUnDA ranged from 1.2 to 5.2 ng/g ww, PFDoDA and PFTrDA ranged from 0.21 to 0.72 ng/g ww, and PFTeDA was below the method detection limits.

A few research and monitoring programs have been carried out to investigate PFASs in freshwater fish in the Arctic, such as in the Canadian Arctic, Norwegian Arctic, Greenland and the Faroe Islands (Ali et al., 2021a; Bossi et al., 2015; Bossi et al., 2005; Butt et al., 2010; Lescord et al., 2015; Norwegian Environment 2013). Typically, C9 - C12 PFCAs predominate in most samples of freshwater fish. Low levels of PFASs were detected in trout samples from Canadian and European Arctic, while the pattern was different elsewhere. It was found that C9–C14 PFCAs predominated in trout from the European Arctic, but they were below the method detection limits (<0.01 ng/g ww) in lake trout from the Canadian Arctic.

PFASs in marine mammals were reviewed by Butt et al. and Muir et al. in 2010 and 2019 (Butt et al., 2010; Muir et al., 2019). PFOS was the predominant PFAS in seals from Greenland and Svalbard, with contributions higher than 60%. The longer odd chain PFASs (PFUnDA, PFNA and PFTrDA) were the most predominant PFCA (Butt et al., 2014; Smythe et al., 2018).

New PFASs have been reported in the liver of ringed seals from northwest Greenland and in hooded seals (Cystophora cristata) from the Greenland Sea (Rotander et al., 2012a), including PFTeDA, PFPeCA and C16-PFCA (Muir et al., 2015). 6:2 Cl-PFESA was detected in ringed seal liver from East Greenland at 0.045 ± 0.004 ng/g ww, and PFECHS was found in ringed seal liver. Polyfluoroalkyl phosphate esters (mono- and diPAPs) in seal liver were below the method detection limit (<0.5 ng/g ww). The C9-C12-PFCAs along with PFOS and PFOSA were the major PFASs in whales from the Canadian and European Arctic, while the detection frequencies were less than 2% for PFOA, PFHpA, and PFHxA.

PFASs have been investigated in polar bears from Greenland and Svalbard. In most studies, PFOS was the predominant PFAS, followed by PFNA and PFUnDA. In East Greenland polar bears, the concentrations of PFSA and PFCA were following the sequence: liver > blood > brain > muscle (Greaves et al., 2013). Boisvert et al. investigated the bioaccumulation and biomagnification of 22 major PFASs in tissues of polar bears (Ursus maritimus) and the ringed seal (Pusa hispida) from East Greenland (Boisvert et al., 2019). C9-C11 PFCAs were dominant in the liver, while C11-C14 PFCAs contributed more to bear fat and seal blubber. The BAFs of C9 – C13 PFCA decreased with increasing chain length. BAFs from seal fat to bear liver reflect dietary exposure relationships to PFASs between bears and seals.

Seabird eggs, liver and blood are typical matrices for watching PFASs in the Arctic (Butt et al., 2007; Lucia et al., 2015; Martin et al., 2004). PFUnDA was found to be the dominant PFCA in thick-billed murre, glaucous gull and black guillemot, while PFTrDA was predominant in black-legged gulls and northern gulls (Braune and Letcher 2013; Braune et al., 2014). PFBS was detected in the murre and fulmar eggs ranging from 0.04 ng/g (murres) to 0.57 ng/g (fulmars) from 2010 to 2011, while PFOS ranged from 20 ng/g (fulmars) to 24 ng/g (murres) (Braune and Letcher 2013). The median concentrations of Σ PFSA and Σ PFCA were 300 and 100 ng/g dw in peregrine falcon eggs in the South Greenland (Vorkamp et al., 2019).

In a marine food web from the coastal area of Longyearbyen (Ali et al., 2021a), the PFAS profiles changed with varying trophic level. High proportions of long-chained PFCA and 6:2 FTS were observed in zooplankton and polychaetas. While linear PFOS dominated in fish and gull liver.

PFBS and 6:2 FTS showed bioaccumulation potential in marine invertebrates, whereas they were not detected in organisms at higher trophic levels.

9.4 Environmental pathways

In polar regions, photodegradation and reaction with OH, Cl or O_3 radicals are the major pathways to eliminate neutral PFASs from the atmosphere (Ellis et al., 2004a; Ellis et al., 2003; Loewen et al., 2005). The PFOA (166 ± 60 pg/L) detected in the ice core during 1977–2015 has been regarded as the background level due to continuous air inputs in the high Arctic (Garnett et al., 2021; Pickard et al., 2018). The accumulation of PFCAs and their precursors in Arctic snow and ice has been observed in Ny-Ålesund and the high Arctic (Garnett et al., 2021; Xie et al., 2015). It is estimated that atmospheric deposition fluxes to the ice cap ranged from 16 to 860 kg/year for PFASs (Young et al., 2007).

In the Antarctic, relatively high levels of PFASs were observed in snow from Dome C(Xie et al., 2020). This result was in line with the high levels of FTOH found in the atmosphere of the Southern Ocean and Antarctic coast. All these analyses suggested that air input might mainly contribute to the trace level of PFSAs in Antarctic. A trace level of 6:2 Cl-PFESA (up to 15.2 pg/L) was detected in lake water in East Antarctic. Moreover, Gebbink et al. observed the presence of 6:2 Cl-PFESA in Greenland marine mammals (Gebbink et al., 2016). As the physicochemical properties of 6:2 Cl-PFESA and PFOS are comparable, transport through oceanic circulation and sea spray is likely to be a dominant transportation pathway.

Marine aerosols or sea spray have been proven to be important media for transporting ionic PFASs over the ocean surface and contribute to LRAT and deposition in the Arctic and Antarctic (Casas et al., 2020; MacInnis et al., 2019; Wolf et al., 2021). Casas et al. reported that the enrichment factors of PFASs in marine aerosols ranged between 522 and 4690 on Livingston Island, Antarctic, which supported the role of marine aerosols as a relevant vector for long-range atmospheric transport of PFASs (Casas et al., 2020).

10 Organophosphate esters

10.1 Introduction

Organophosphorus esters (OPEs) have been applied on a large scale as flame retardants, plasticizers and additives in industrial production, household consumer products and personal care products (Marklund et al., 2005; van der Veen and de Boer 2012). OPEs are continually produced in large quantities since formerly commonly used PBDE flame retardants were prohibited (Wang et al., 2020c). The global production of OPEs was approximately 680,000 tons in 2015 and increased to 1 million tons in 2020 (He et al., 2020). As OPEs are physically added to polymers, they can leach out and be released into the surrounding environment through volatilization, abrasion, and/or dissolution (Brommer et al., 2012; Gravel et al., 2019). Consequently, OPEs have been measured in various environmental matrices, including air, water, sediment and organisms (Fu et al., 2021; Wei et al., 2015). Laboratory tests have shown some OPEs, such as TPhP and TnBP, have toxic effects on metabolic processes and the endocrine system (Ali et al., 2012; EC 2009; Dishaw et al., 2011; van der Veen and de Boer 2012; Waaijers et al., 2013; Wei et al., 2015). Consequently, OPEs have received considerable attention in international studies (van der Veen and de Boer 2012).

The wide presence of OPEs in remote oceans and polar regions has shown that OPEs can undergo LRET via ocean currents and the atmosphere (Castro-Jimenez et al., 2014; Cheng et al., 2013b; Li et al., 2017; Moeller et al., 2012; Moeller et al., 2011c; Salamova et al., 2014a; Salamova et al., 2014b; Suhring et al., 2021; Suhring et al., 2016). As a result, OPEs in the polar regions have attracted great attention from international society and indicate that OPEs are novel organic contaminants and require further study and monitoring to understand their biogeochemistry cycle and toxic impacts in the polar regions (Fu et al., 2021; Xie et al., 2022).

In this work, we review the studies on OPEs in polar regions by focusing on their occurrence spatial distributions in various environmental matrices in the Arctic and Antarctic and their environmental pathways. The most frequently targeted and detected OPEs in the polar regions involved in the review are summarized in Table 18 (Xie et al., 2022).

10.2 Sources and long-range environmental transport

OPEs can be released into the ambient environment during the production, application, and disposal of OPE-related materials (van der Veen and de Boer 2012). As most OPEs have relatively high solubility in water and are difficult to remove by sewage sludge (Bester 2005; Meyer and Bester 2004), they can enter the marine environment through riverine discharge (Schmidt et al., 2020; Wang et al., 2015a; Wolschke et al., 2018). Ocean currents can transport OPEs farther to the remote ocean and reach polar regions (Li et al., 2017; Na et al., 2020b; Suhring et al., 2021). OPEs can be released into the air from OPE-containing materials and households, especially from electric waste treatment plants (Liu et al., 2016; Nguyen et al., 2019; Zeng et al., 2020). It was estimated that the atmospheric half-life of OPEs increased from <15.3 h in the gas phase to 3.5–14 days in the particle phase, suggesting that OPEs can be transported to long distances via the atmosphere (Liu et al., 2014). Many studies have revealed the presence of OPEs in remote environments, including ocean and polar regions, implying their long-range transport potential (Castro-Jimenez et al., 2016; Cheng et al., 2013b; Chokwe et al., 2020; Moeller et al., 2012; Na et al., 2020b). Model studies have shown that chlorinated OPEs can be efficiently transported via ocean currents due to their persistence, lower volatility and high solubility (Rodgers et al., 2018; Sühring et al., 2016). Thus, both ocean currents and the atmosphere are significant media for transporting OPEs from the source area to the polar regions.

Table 18:	The names, abbreviations, chemical formulas and physicochemical properties of
	the most regularly detected OPEs in polar regions

Compound	Abbreviation	CAS No.	Solubility ^b (mg/L, 25°C)	VP ^{a,b} (Pa, 25 °C)	logKow ^b
Tris (2-chloroethyl) phosphate	ТСЕР	115-96-8	878	8.17	1.44
Tris (1-chloro-2-propyl) phosphate	TCIPP	13674-84-5	51.8	7.53×10 ⁻³	2.59
Tris (1,3-dichloro-2-propyl) phosphate	TDCIPP	13674-87-8	1.50	3.81×10 ⁻⁵	3.65
Trimethyl phosphate	тмр	512-56-1	3.0×10 ⁵	55.3	-0.65
Triethyl phosphate	ТЕР	78-40-0	1.1×10 ⁴	22	0.80
Tripropyl phosphate	TnPP	513-08-6	827	3.08	1.87
Tri-isopropyl phosphate	TiPrP	513-02-0	501	18.4	2.12
Tributyl phosphate	TnBP	126-73-8	280	0.151	4.00
Tri-isobutyl phosphate	TiBP	126-71-6	16.2	1.71	3.60
Tripentyl phosphate	TPeP	2528-38-3	0.332	2.33×10 ⁻³	5.29
Trihexyl phosphate	тнр	2528-39-4	0.0102	3.29×10 ⁻⁴	6.76
Triphenyl phosphate	трнр	115-86-6	1.9	1.49×10 ⁻³	4.59
Tris(2-butoxyethyl) phosphate	ТВОЕР	78-51-3	1.96	1.65×10 ⁻⁴	3.75
Tris(2-ethylhexyl) phosphate	тенр	78-42-2	1.46×10 ⁻⁵	1.10×10 ⁻⁵	9.49
2-ethylhexyl diphenyl phosphate	EHDPP	1241-94-7	0.0666	4.45×10 ⁻³	5.73
Tricresyl phosphate	TCrP	1330-78-5	0.207	1.62	5.11
Di-n-octylphenyl phosphate	DOPP	6161-81-5	4.25×10 ⁻⁴	9.88×10 ⁻⁶	8.04
Methyl diphenyl phosphate	MDPP	115-89-9	61.6	1.55×10 ⁻³	2.93

a Vapor pressure

b Solubility, $V_{\rm p},\, logK_{\rm ow}$ are adopted from EPIWEB 4.1. Source: Xie et al. 2022

10.3 Occurrences in environmental matrices

10.3.1 OPEs in air

OPEs have been widely detected in the atmosphere of the Arctic, the Antarctic and the Southern Ocean. The concentrations of \sum_{8} OPEs ranged from 120 to 2900 pg/m³ in particles over the Pacific, Indian, Arctic, and Southern Oceans in 2010-2011(Moeller et al., 2012) and a few to hundreds of pg/m³ in the particle samples collected in the West Pacific, the Indian Ocean and the Southern Ocean over 2009-2010 (Cheng et al., 2013b), suggesting their circumpolar and global distribution.

Sühring et al. found a median concentration of 50 pg/m³ for Σ_{13} OPEs in the Canadian Arctic from 2007 to 2013 (Sühring et al., 2016), and Salamova et al. reported Σ_{13} OPEs in particles at concentrations up to 1450 pg/m³ (median: 330 pg/m³) in Longyearbyen from 2012 to 2013 (Salamova et al., 2014a). OPEs in the gas phase were measured in passive air samples ranging from 360-850 pg/m³ at Ny-alesund (Han et al., 2020). Furthermore, OPEs have been measured in both gas and particle phases, with concentrations from 35 to 340 pg/m³ in the European Arctic (Li et al., 2017), 230 to 1900 pg/m³ in the Northwestern Pacific to the Arctic Ocean(Na et al., 2020b), and 74 to 460 pg/m³ (mean: 200 pg/m³) at Zeppelin in 2020 (Bohlin-Nizzetto et al., 2021) (Figure 12).



Figure 12: Mean concentrations of OPEs in gas (G) and particle (P) phases in the Arctic, Antarctic and oceans

Source: Fu et al. 2021

In the Antarctic, the concentrations of OPEs ranged from 6.0 to 140 pg/m³ in ship-bound particle samples (Cheng et al., 2013b) and from 34 to 400 pg/m³ (mean: $120 \pm 12 \text{ pg/m}^3$) in the western Antarctic Peninsula (Wang et al., 2020). Among the OPEs, chlorinated OPEs such as TCEP and TPCIPP are the predominant compounds, and TnBP and TiBP are the major non-chlorinated OPEs.

10.3.2 OPEs in seawater, snow and ice

The OECD POV and LRTP screening tool predicted that OPEs tend to distribute in water, suggesting that ocean currents are an important medium for the LRET of OPEs (Rodgers et al., 2018; Sühring et al., 2016). The concentrations of OPEs ranged from 0.35 to 8.4 ng/L (\sum_{8} OPEs)

in the North Atlantic to the Arctic (Li et al., 2017) and from 8.5 to 140 ng/L (\sum_{11} OPEs) from the North Pacific to the Arctic (Na et al., 2020b). Suehring et al. detected 11 OPEs in surface water from 2013-2018 in the Canadian Arctic, with mean concentrations of 10 ng/L and 1.3 ng/L for Σ chlorinated OPEs (Cl-OPEs) and Σ non-chlorinated OPEs, respectively (Suhring et al., 2021). Comparable OPE concentrations were measured with a passive water sampler (Gao et al., 2019; McDonough et al., 2018) (Figure 13). Interestingly, relatively high concentrations of OPEs were reported in seawater ranging from 8.7 to 360 ng/L (mean: 89 ng/L) in Ny-alesund, Arctic (Gao et al., 2020), from <5.0 to 44 ng/L Σ_6 OPEs in seawater of Fildes Peninsula (Gao et al., 2018), and 20-9200 ng/L in freshwater from the northern Antarctic Peninsula (Esteban et al., 2016). Such high levels are feasible only due to local sources, such as discharge of effluents of the research stations (Choi et al., 2020).



Figure 13: Mean concentrations of OPEs (pg/L) in river water, lake water and seawater in the Arctic and Antarctic

Source: Fu et al. 2021

OPEs have been investigated in surface snow in the Arctic and Antarctic. TCEP was detected in surface snow on the Antarctic ice sheet, with concentrations ranging from 0.05 to 2.0 ng/L (Cheng et al., 2013a). Xie et al. reported the occurrence of OPEs in snow samples collected at Dome C, with total concentrations ranging from 7.2 to 20 ng/L (Xie et al., 2020) (Figure 14), which was in line with the levels detected in Arctic snow (4.4 to 11 ng/L, mean: 7.8 ng/L) (Li et al., 2017) and in Canadian Arctic Lake Hazen and its tributaries (15–25 ng/L) (Sun et al., 2020). These pioneering works showed that LRAT and snow precipitation play important roles in the occurrence and distribution of OPEs in polar regions.



Figure 14: Concentrations of 9 OPEs in snow samples from Dome C

Source: Xie et al. 2020

10.3.3 OPEs in sediment

OPEs have been investigated in sediment from the Arctic Ocean (Table 19). The concentrations of Σ OPEs ranged from 0.16 to 4.7 ng/g dw from the North Pacific to the Arctic Ocean, (Ma et al., 2017), 0.01–15 ng/g dw in Ny-Ålesund, Svalbard (Gao et al., 2020), and with a concentration ranging from 0.12 to 57 ng/g dw and at a median of 8.3 ng/g dw in the Canadian Arctic (Suhring et al., 2021). Chlorinated OPEs (especially TCEP and TCIPP) have been the most abundant detected OPEs in ocean sediments. Generally, the extent of sediments and the water column as a final sink of OPEs will depend on the water column biogeochemistry, which needs further research.

10.3.4 OPEs in biota

Concentrations of OPEs are significantly lower in birds and mammals than in fish. For example, one study investigating OPEs in 8 species in Svalbard, Norway, reported that 10 OPEs were found in capelin, while no individual OPEs were found in most of the other species, including kittiwake, Brünnich's guillemot, glaucous gull, ringed seal, harbor seal, arctic fox and polar bear (Hallanger et al., 2015). Five out of 17 OPEs were detectable at a few ng/g in polar bear fat samples with low detection frequencies (Letcher et al., 2018).

The bioaccumulation and biomagnification potentials of OPEs in marine organisms depend on their physicochemical properties, bioavailability and extent of biotransformation (Kim et al., 2011). In the occurrence of OPEs in Arctic biota, even lower detection frequencies (DFs) and

concentrations of OPEs were observed in higher trophic levels (such as birds, seals, arctic foxes and polar bears) than in fish (Hallanger et al., 2015). This may be due to either poor assimilation from the diet or rapid metabolism of OPEs in these species, such as birds and polar bears (Letcher et al., 2018). More attention should be given to the bioaccumulation behavior of OPEs in marine organisms to determine their biomagnification potential.

Location	Ny-Ålesund	North Pacific and Arctic Ocean	Bering Sea	Bering Strait	Chukchi Sea	Canadian Basin	Central Arctic Ocean
ТСЕР	<0.02-2.9	0.54	0.66	0.11	0.28	0.72	1.1
ТСІРР	0.01-7.4	0.068	0.081	0.009	0.033	0.11	0.14
TDCIPP	<0.02-0.73	0.014	0.011	nd	0.006	0.037	0.028
TnBP		0.068	0.11	0.06	0.052	0.095	0.061
TiBP		0.16	0.32	0.17	0.13	0.18	0.12
TPeP		0.007	0.002	0.001	0.002	0.02	0.015
TBOEP	<0.02-2.6						
трнр	<0.01-0.37	0.023	0.03	0.001	0.023	0.028	0.028
TCrP	<0.01-0.64						
EHDPP	<0.01-0.87						
ΣΟΡΕs	0.01-15	0.16-4.7	1.2	0.35	0.52	1.2	1.5

Table 19:Occurrence of OPEs (ng/g dw) in surface sediment in the Arctic

Source: Gao et al. 2020, Ma et al. 2017

10.4 Environmental pathways

The air-seawater diffusive exchange of OPEs in the Arctic oceans interferes with the LRET of OPEs. TCIPP and TCEP exhibited net deposition fluxes from the North Pacific to the high Arctic (Na et al., 2020b), while net volatilization flux controlled the air-water exchange for OPEs in the European Arctic (Li et al., 2017). McDonough et al. found OPEs at dynamic equilibrium across the Farm Strait in the Arctic, with the exception of some volatilization of TnBP observed at Cape Bounty Lake sites in 2016, of TDCIPP in the Barrow Strait in 2015, and the deposition of TPHP (McDonough et al., 2018). Atmospheric particle deposition is also an important input for OPEs in polar regions. The dry particle-bound OPE deposition fluxes ranged from 14 to 94 ng/m²/day in the Northeast Pacific and the Arctic (Na et al., 2020b) and from 2-16 ng/m²/day in the European Arctic (Li et al., 2017).

In polar regions, snow and rain act as effective scavengers for atmospheric OPEs in the Arctic, the Southern Ocean and the Antarctic (Casal et al., 2019). The high OPE concentrations found in the snow samples from the Arctic and Antarctic expeditions showed clear evidence of an input source to the polar environment (Cheng et al., 2013a; Li et al., 2017; Xie et al., 2020). Along with the processes of ice retreat and snow melting, OPEs accumulated in snow could be directly released into the water column (Li et al., 2017; Na et al., 2020b; Sun et al., 2020).

11 Pharmaceuticals and personal care products (PPCPs)

11.1 Introduction

PPCPs include all chemicals used for health care, cosmetics and medical purposes. There are currently more than 3000 PPCPs on the market, and new molecules enter the market every year (Arpin-Pont et al., 2016). PPCPs enter the environment indirectly through wastewater from households or agriculture (livestock). Since their removal efficiency from wastewater is not high and because most compounds are not degraded or are only slowly degraded, they reach the aquatic environment through wastewater discharge. Some personal care products (PCPs), such as UV filters in sunscreens, synthetic fragrance materials, and volatile methyl siloxanes (VMS), can enter the environment directly through recreational activities. This group of compounds can generally be considered "pseudo-persistent" because their degradation rate is slow relative to a large amount of quantitative input/discharge into the environment (Daughton 2003). Wide range of medicinal products available for human or veterinary use may cause global environmental problems (Klatte et al., 2017).

Cyclic volatile methyl siloxanes (cVMS), such as octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6), have been applied as ingredients in PCPs and can be released to aquatic environments through wastewater effluents (Krogseth et al., 2017). 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) Nevertheless, their fate has become an environmental issue. Transport of PPCPs to polar regions via ocean currents remains unclear as only limited data are available on the occurrence of various drugs in coastal environments (Arpin-Pont et al., 2016). Several national and international research programs have been carried out to investigate PPCPs in various matrices in the Nordic environment. In 2016, the AMAP conducted a comprehensive survey of the occurrence and fate of PPCPs in the Arctic, which raised concerns about PPCPs in polar regions.

11.2 Sources and long-range environmental transport

In polar environments, pharmaceutical residues are released mainly into aqueous environments through wastewater discharge, which often has had little or no treatment applied (Esteban et al., 2016). Low biodegradability would be expected for the majority of PPCPs entering the aquatic system. Therefore, the environmental toxicological consequences of sustained releases of PPCP in polar regions are expected to differ from those in temperate regions of the world. The availability of PPCP data in the polar regions is more limited than that of temperate ocean systems. Kallenborn et al. reported that the presence of PPCPs in the Arctic was related to local sources. Over 100 PPCP-related compounds have been identified in various Arctic environmental matrices, from territorial waters to high trophic biota (Kallenborn et al., 2018). The majority of PPCPs were determined in the influent, effluent, and sludge from sewage. Residues of certain PPCPs were also reported in river water, seawater, sediment, marine fish and mammals in the circulate environment of polar regions. Some volatile and semi-volatile PPCPs have also been measured in air.

11.3 Occurrences in environmental matrices

11.3.1 PPCPs in air

Among PPCPs, limited data are available for UV-Fs and FMs in air. HHCB and AHTN were the two most abundant FMs in 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). Some UV-Fs, such as homosalate (HMS), 2-ethylhexyl salicylate (EHS) and 2-ethylhexyl 4-methoxycinnamate (EHMC), have been detected in both gaseous and particle phases in urban air in Canada (Pegoraro et al., 2020). In addition, UV-Fs were detected in streams formed by melting snow and ice.

As typical volatile compounds, CVMS was measured in air collected at Zeppelin station in 2011 (Krogseth et al., 2013). The average concentrations in air were 2.9 ± 0.46 and 0.45 ± 0.18 ng/m³ for D5 and D6, respectively, and ranged from n.d. to 2.2 ng/m³ and n.d. to 0.95 ng/m³ in summer and from n.d. to 3.0 ng/m³ and n.d. to 2.1 ng/m³ in winter, respectively. Half-live times (t¹/₂) were estimated as 10 , 6.7, and 5 d for D4, D5, and D6, respectively, which are greater than the criterion of 2 d for LRT substances. Obviously, more studies on UV-Fs and FMs in air from polar regions are required to reveal their LRAT.

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Location	Atenolol (ng/L)	Carbamazepine (ng/L)	Metoprolol (ng/L)	Naproxen (ng/L)
lqaluit, NU	nd – 410	nd – 110	nd – 510	nd – 5200
Baker Lake, NU	nd–50	nd–550	nd–90	nd–550
Cambridge Bay, NU	nd – 90	nd–2700	nd–170	nd–360
Kugluktuk, NU	45–270	510–1800	52–400	420–1100
Cambridge Bay, NU	nd–97	1.2–430	nd	nd
Tromso, Norway	-	nd–270	nd–340	-
Klaksvic, Faroe	nd	-	2.82	nd
Tórshavn, Faroe	nd–13	-	nd–650	nd–1800
Akureyri, Iceland	501–710	-	14–95	520-2300
Hveragerði, Iceland	980–2200	-	66–160	180–190
Reykjavík, Iceland	1000-1600	-	98	1200-2300

Table 20:	Concentrations of four pharmaceuticals in effluents of different Arctic wastewater
	systems

nd: not detected

Source: Stroski et al. 2020, Weigel et al. 2004, Huber et al. 2016, Chaves-Barquero et al. 2016.

11.3.2 PPCPs in river water, lake water and seawater

PPCPs have been included in the Nordic environmental screening program for inspection of their occurrences in seven Nordic countries in Europe. More than 50 PPCPs were found in both the influent and the effluent of sewage with concentrations from a few ng/L to μ g/L (Kallenborn et al., 2018). Apart from the original substance of PPCPs, metabolites and transformed products of PPCPs were identified in sewage effluent. For example, carboxy-ibuprofen, one of the ibuprofen metabolites, was measured up to 70 μ g/L in Tromsø and 38 μ g/L in Longyearbyen,

respectively (Kallenborn et al., 2018; Weigel et al., 2004). Stroski et al. reported 7 PPCPs (atenolol, carbamazepine, metoprolol, naproxen, sulfapyridine, sulfamethoxazole, and trimethoprim) in wastewater effluents from four Canadian Arctic communities(Stroski et al., 2020). Together with the results from the studies in the European Arctic, naproxen (nd - 5200 ng/L) was the predominant PPCP released from WWTPs, followed by carbamazepine (nd-2700 ng/L), metoprolol (nd-650 ng/L) and atenolol (nd-2200 ng/L) (Figure 15 and Table 20).

Two FMs, HHCB and AHTN, were measured in the European Arctic with mean concentrations of 59 and 23 pg/L in seawater (Xie et al., 2007). Tsui et al. reported that BP-3, EHMC and OC were the dominant FMs in seawater from the Arctic, with concentrations of 17, 25 and 26 ng/L, respectively. Generally, the levels of UV-Fs in the Arctic were 1-2 orders of magnitude lower than those from the urban environment (Tsui et al., 2014). Vecchiato et al. investigated fragrance materials (FMs) in seawater and snow collected in Ny-Ålesund, Svalbard (Vecchiato et al., 2018). Concentrations of FMs up to 72 ng/L were detected in the surface snow from the town to the glacier, indicating the LRAT of FMs.

A range of potentially toxic contaminants have been determined, including UV filters, parabens, antimicrobial compounds and pharmaceutical residues, in the wastewater effluent of research bases in the Antarctic (Emnet et al., 2015; Esteban et al., 2016). The concentrations of methyl-triclosan, 4-methyl-benzylidene camphor, octylphenol, benzophenone-1, and octylphenol were comparable to the levels in the urban environment. PPCPs such as alkylphenol, parabens and UV filters were detectable in seawater at distances up to 25 km from the research stations. Sewage discharge and disposal of raw human waste contributed major PPCPs in the Antarctic coastal environment. Alkylphenol (1.1–7200 ng/L) was measured in surface water, including streams, ponds, glacier drains, and urban wastewater discharge into the sea from the Northern Antarctic Peninsula region. Domínguez-Morueco et al. further evaluated the presence of UV-Fs in both dissolved and particle phases in wastewater and freshwater in the northern Antarctic Peninsula region (Dominguez-Morueco et al., 2021). UV-Fs ranged from <LOD to 1300 ng/L in water and from LOQ to 33 μ g/g dw in the particle phase. BP-1 and BP-3 were the dominant UV filters in wastewater, but 4-MBC was quantified in stream water and pond water with concentrations ranging from 18-45 ng/L.

Gonzalez-Alonso et al. analyzed 44 PPCPs in water samples from streams, ponds, glacier drains, and a wastewater discharge into the sea in the northern Antarctic Peninsula region (Gonzalez-Alonso et al., 2017). Acetaminophen (49 µg/L), diclofenac (15 µg/L), ibuprofen (10 µg/L), and the stimulant caffeine (71 µg/L) were the dominant PPCPs in waters discharged directly into the ocean without any prior purification processes. Eight antibiotics were detected in treated wastewater from King George Island and mainly included the quinolones ciprofloxacin (92% samples, 0.89 µg/L) and norfloxacin (54% samples, 0.75 µg/L), the macrolides azithromycin and clarithromycin (15% samples, 0.4 µg/L), and erythromycin (38% samples, 0.003 µg/L), metronidazole (1 sample, 0.17 µg/L), clindamycin (1 sample, 0.1 µg/L) and trimethoprim (2 sample, 0.001 µg/L) (Hernandez et al., 2019). Ciprofloxacin was detected in seawater at concentrations ranging from 4 to 218 ng/L, suggesting discharge from wastewater to the coastal environment. Bacterial resistance was also observed for some antibiotics (such as trimetropim and nalidixic acid) identified in the sample.

Fragrance materials, such as ambrofix, amylicylate, benzyl salicylate, hexyl salicylate, lemonile and lkoumal, were detected in the coastal surface seawater of Terra Nova Bay in the Ross Sea, Antarctic, with total concentrations up to 100 ng/L (Vecchiato et al., 2017). The Italian research station Mario Zucchelli (MZS) could be a primary source discharging FMs into coastal water. However, the increasing concentrations measured in the seasonal meltwater implied reemissions of FMs from sea ice and snow in the Antarctic.





Source: Chaves-Barquero et al. 2016, Huber et al. 2016, Stroski et al. 2020, Weigel et al. 2004

11.3.3 PPCPs in organisms

Warner et al. investigated cVMS in sediment and biota in the European Arctic (Warner et al., 2010). High concentrations were meaured for D5 in fish, with averages of 180 and 530 ng/g lw in Atlantic cod and shorthorn sculpin, while relatively low concentrations of D5 $(1.9 \pm 0.2 \text{ ng/g} \text{ lw})$ and D6 (0.8-1.1 ng/g lw) were found in seal blubber from Kongsfjorden (Table 21). The cVMS were measured from Lake Storvannet in Northern Norway, and the concentrations of D5 were 60 ± 1.2 in Chironomidae larvae, 110 ± 4.5 in pea clam *Pisidium* sp., 130 ± 100 in sticklebacks, 41 ± 38 in char, and 9.9 ± 5.9 (trout) ng/g ww (Table 21). VMSs have also been detected in soils, vegetation, phytoplankton, and krill from the Antarctic Peninsula region (Sanchis et al., 2015). D4 and D5 were the predominant compounds, with concentrations from MDLs to 110 ng/g dw in soil. Concentrations of cVMSs in phytoplankton were negatively correlated with salinity, suggesting input from melting ice and snow. UV filters, parabens and alkylphenol were detected in biota from coastal areas near Antarctic research stations (Emnet et al., 2015). There are very few data regarding the presence of pharmaceuticals in biota. Given their high concentrations in effluents and coastal water, more studies on the bioaccumulation of certain PPCPs in marine fish and mammals are needed.

Sample	Location	D3	D4	D5	D6
Soil (ng/g dw)	Antarctic	16	14	33	22
Vegetation (ng/g dw)	Antarctic	1.6	6.2	18	32
Phytoplankton (ng/g dw)	Antarctic	4.2	0.93	3.2	1.2
Krill (ng/g dw)	Antarctic	36	49	37	35
Sculpin (ng/g lw)	Svalbard	0	<2.2	270	7.8
Atlantic cod (ng/g lw)	Svalbard	0	<2.2	97	18
Brown trout (ng/g ww)	Lake Storvanet			9.9 ± 5.9	
Arctic char (ng/g ww)	Lake Storvanet			41 ± 38	
Sticklebacks (ng/g ww)	Lake Storvanet		13 ± 9.2	130 ± 100	6.3 ± 3.6
Chironomidae (ng/g ww)	Lake Storvanet		9.9 ± 0.3	60 ± 1.2	9.3 ± 0.1
Pisidium sp. (ng/g ww)	Lake Storvanet		4.7 ± 0.4	110 ± 4.5	12 ± 1.2
Arctic Char (ng/g)	Svalbard		<3.1	<2.2	<10

Table 21:	VMS Concentrations in biota in the Arctic, and in soil, vegetation, phytoplankton
	and krill in the Antarctic

Source: Sanchis et al. 2015, Krogseth et al. 2013, Warner et al. 2010, Krogseth et al. 2017, Lucia et al. 2015.

11.4 Environmental pathways

The occurrence and distribution of PPCPs in the Arctic and Antarctic suggested that both oceanic currents and the atmosphere might transport PPCPs to polar regions. 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. Thus, wastewater runoff could be an important local contamination source of PPCPs. 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 Antarctic. This is considered important when large amounts of antibiotics/antimicrobial agents are released to increase the possibility of developing resistance (Kallenborn et al., 2018). Antibiotic resistance (AR) genes have been found in sediment, soil and water in the Arctic (Hayward et al., 2018; Makowska et al., 2020; Tan et al., 2018). Bacterial resistance was observed for some antibiotics identified in the samples in

sea water and wastewater collected on King George Island (Hernandez et al., 2019). The occurrence of AR in the polar environment could be addressed to the coastal runoff of AR bacteria from terrestrial sources, anthropogenic antibiotic runoff, and selection for resistance in response to antibiotic production in the polar regions (Hatosy and Martiny 2015; Schloss et al., 2010).

Pharmaceuticals usually occur near local sources and have low environmental transport potential. The presence of some PCPs in the ice core and snow could be attributed to LRET, atmospheric deposition and cold condensation in the polar regions (Vecchiato et al., 2020). FMs were detected in ice cores from Elbrus ranging from 130 to 280 ng/L and 72 ng/L in Arctic snow near Ny-alesund. In Terra Nova Bay, Antarctic, increasing FM concentrations were measured in seawater during the seasonal melting of sea ice and snow. In the European Arctic, the deposition fluxes were 1.5 and 5.7 ng/m²/day for HHCB and AHTN, respectively, suggesting that atmospheric input controls the levels of HHCB and AHTN in the polar region. These studies indicated that FMs could reach the polar regions via LRT (Vecchiato et al., 2017).

12 Summary, conclusions and future perspectives

12.1 Summary of the key findings

This report reviewed studies on the occurrence of EOCs in both the Arctic and the Antarctic. The long-range environmental transport processes of EOCs via ocean currents and the atmosphere from continental sources to polar regions were evaluated. Literature data showed that environmental pathways such as atmospheric deposition, air-water exchange and discharge from seasonal melting could interfere with the biogeochemical cycle of EOCs in polar regions. Local emissions of certain EOCs may occur near research stations, harbor and tourism sites. The major findings for ten groups of EOCs are given as follows.

POPs such as OCPs, PCBs, PBDEs, PCNs, dioxin and PAHs have been systematically monitored in air at several Arctic stations and have shown seasonal variations and declining trends. These chemicals were also the major POPs reported in the Antarctic, while their concentrations were considered lower than those obtained from Arctic stations, illustrating the combination of remoteness and lack of potential local sources for Antarctic. Model simulations showed the influence of continental air on the transport of POPs.

EOCs, such as PFASs, OPEs, CPs and PPCPs, were investigated in air, water, snow, ice and organisms in the Arctic. Selected EOCs (PFASs and CPs) have been included in long-term atmospheric monitoring programs at Ny-alseund, Nord station in Greenland, and Alert, Canada. Screening surveys for EOCs in environmental and biological matrices have been carried out through national and regional research programs. Data on EOCs in Antarctic are rare. As most studies were carried out in summer near the research station, local emissions released from the stations and human activities may interfere with the measurements, which can lead to overestimations of the background levels of EOCs. Nevertheless, the LRET of certain EOCs has been highlighted with their occurrences in ice cores, snow and lake waters. Other emerging chemicals, such as current-use pesticides, volatile PFASs, alternative BFRs, hexabromcyclododecane (HBCD), tetrabrombisphenol A (TBBPA) and its derivatives, phthalate esters and microplastics have also been reported in the polar regions.

Observations at research stations in the Arctic and Antarctic have shown temporal trends of class POPs and spotted the occurrence of EOCs. Geographic distributions highlighted different contamination sources and influences from meteorological conditions. Ship-bound air, water and sediment samples along the Pacific and Atlantic transects have revealed LRET from continental sources to the polar regions via both ocean currents and the atmosphere. Air-water gas exchange and atmospheric particle deposition control the mass balance of POPs and EOCs in the transition area of the polar regions. In addition, abundant snow precipitation is an important pathway for the input of EOCs in the polar region. Consequently, snow and ice pack become an intermediate sink for EOCs and can release accumulated chemicals to the air and ocean during the melting process.

LRAT is the primary pathway that conveys POPs from continents to polar regions. The Arctic is surrounded by countries with dense populations and advanced industrialization. High emissions released from these regions can be quickly transported to the Arctic. Many studies have shown that the contamination levels of POPs are higher in the Arctic than in the Antarctic.

In the Northern Hemisphere, polar EOCs such as ionic PFAS, OPEs and PPCPs could be transported via oceanic currents (North Pacific and Atlantic) to the Arctic Ocean, which has been evidenced by the latitudinal distribution from low latitudes to high Arctic oceans. In the Southern Hemisphere, the Antarctic is remote from any continent and surrounded by the

Southern Ocean. The Antarctic Circumpolar Current forms a physical barrier inhibiting the effective north–south transport of surface waters, which limits the oceanic transport for EOCs, e.g., PFASs and PPCPs, to the Antarctic. However, the barrier has a gap in which some exchange occurs through upwelling of deeper and older waters. Therefore, EOCs defused in deeper water can break through the barrier and reach the Southern Ocean and the Antarctic. The concentrations of EOCs are usually 1-2 orders of magnitude lower than those present in the Arctic Ocean.

Increasing human activity, such as polar expedition, shipping and tourism in the Arctic and Antarctic, represents a potential direct source of EOCs. Although human activities in Antarctic are limited to scientific research and tourism is subject to many restrictions, the steady increase in human activities seems to have a significant impact on terrestrial and aquatic ecosystems, especially marine ecosystems. Studies of PPCPs, ionic PFASs and currently used plasticizers and flame retardants in the effluents and coastal water from the research stations show clear evidence that the research stations and local settlements act as local sources for EOCs.

Meanwhile, an assessment of the interaction between Arctic climate change and POPs and chemicals of emerging Arctic concern was carried out by the AMAP in 2019-2021 under unique changes in Arctic snow, water, ice and permafrost conditions (AMAP 2021). A perturbed air-surface coupled model was developed to simulate and predict perturbations of POP concentrations in various environmental media under given climate change scenarios. OCPs and PCBs showed a strong response to specified climate change scenarios. More studies have shown that glacier ice and snow act as secondary emission sources in polar regions and release POPs and EOCs into the atmosphere and ocean.

Various studies have reported relatively high concentrations of POPs and EOCs in migratory biota, marine mammals, and fish in polar regions. Bioaccumulation and magnification have been well studied for classic POPs. Despite only a few data being available for EOCs, some EOCs have shown high bioaccumulation potential. The marine species living downstream of the research stations and local settlements may face high pressure from local emissions, such as PPCPs and PFASs.

12.2 Conclusions and perspectives

Legacy POPs in environmental media and biota have shown declining temporal trends in the Arctic and the Antarctic during the global attempt to eliminate their manufacture and application. However, the previous inventory of legacy POPs in the environment is likely to enter the global cycle again because of climate change. Thus, continual monitoring of the temporal variability of legacy POPs in environmental matrices is needed.

EOCs are widely present in various environmental matrices and organisms in the Arctic and Antarctic. Studies in the Arctic showed that EOCs are the main chemical group in both the environment and the biota and exceeded 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 expedition and tourism in the polar region can directly contribute to the local environment. Therefore, background measurements of EOCs need to be considered to minimize the interferences. Current research mostly focuses on oceanic transects and aquatic environments. Comprehensive surveys and long-term monitoring of EOCs in air, water and terrestrial biota in the Arctic and Antarctic need to be strengthened to elucidate their LRET, persistence, bioaccumulation and toxicity (Fu et al., 2021).

The transportation pathways of EOCs to the Arctic and the Antarctic have not been well understood due to Lack of observation in both spatial and time. Many models have been established to predict the fate and transport of POPs from source regions to polar areas and have attempted 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 freshwater (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 airwater/snow exchange. In addition, accurate and high-resolution data of EOCs are required to improve model predictions for their environmental fates and health impact.

Concentration thresholds need to be established for individual compounds as well as for actual cocktail mixtures that actually show biologically relevant but not yet statistically determined relationships. 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 climate change should be explored to assess the actual health risks to humans and animals in polar regions (Dietz et al., 2019; Letcher et al., 2010; Szopińska et al., 2019).

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

12.3 Recommendations

- Long-term monitoring programs of POPs at research bases need to be continued for air and precipitation. EOCs and transformation products should be considered in monitoring their physicochemical behavior.
- The occurrence, bioaccumulation and biomagnification of POPs and EOCs in territory species and marine organisms in the Arctic and Antarctic should be extensively studied.
- Laboratory tests need to be performed to establish concentration thresholds for individual compounds as well as for realistic cocktail mixtures of EOCs that in fact indicate biological relevance.
- A sample bank for both environmental matrices and biological species should be initiated through national and international programs for a retrospective analysis to assess alternative EOCs in the future.
- Nontargeted screening approaches using high-resolution mass spectrometry coupled to liquid or gas chromatography have arisen as new approaches in analytical chemistry to identify and eventually quantify novel emerging chemicals.
- Metrological and atmospheric chemistry observations should be established along with monitoring programs. Accurate precipitation rates (snow and rain) need to be recorded as well. These parameters are required to estimate environmental fate and exchange fluxes between different spheres in polar regions.

- Model development is toward prediction for the impact of climate change on the levels, temporal trends, transportation and bioaccumulations of POPs and EOCs in the polar regions. Thus, high-resolution oceanography, sea ice, snow, and freshwater data in the Southern Ocean and Arctic are requested.
- Comprehensive observation coupled with multicompartment models could be used to improve the understanding of the health impact of POPs and EOCs on organisms and human beings in polar regions.
- Climate change directly and indirectly impacts the sources, transport pathways, and fate
 of POPs and EOCs (AMAP 2021). Reemissions of legacy POPs and EOCs accumulated in
 glaciers, sea ice, and snow may alter the concentrations and amplify their effects in polar
 regions. Therefore, future research needs to understand the various biogeochemical and
 geophysical processes under climate change and anthropogenic stress in order to be able
 to predict the environmental fate and toxicity risk of EOCs in polar regions.

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A Appendix

No.	Compounds	logK _{ow} (observed)	logK _{ow} (predicted)	SE-Pred.	Difference ^b	logK _{ow}
3	4-MonoBDE		4.31	±0.19		4.96 ^e
7	2,4-DiBDE		5.32	±0.13		5.39 ^e
8	2,40-DiBDE		5.28	±0.13		5.45 ^e
15	4,40-DiBDE		4.84	±0.16	0.05	5.51 ^e
17	2,20,4-TriBDE	5.74 ± 0.22ª	5.79	±0.10	0.02	5.53 ^d
28	2,4,40-TriBDE	5.94 ± 0.15ª	5.96	±0.09		5.53 ^d
30	2,4,6-TriBDE		6.12	±0.09		5.8 ^c , 5.84 ^e
32	2,40,6-TriBDE		5.89	±0.10		5.8 ^c , 5.80 ^e
35	3,30,4-TriBDE		5.57	±0.11		5.8 ^c , 5.87 ^e
47	2,20,4,40-TeBDE	6.81 ± 0.08ª	6.76	±0.06	0.05	6.2 ^c , 6.02 ^d , 6.26 ^e
77	3,30,4,40-TeBDE		5.84	±0.10		6.2 ^c , 6.42 ^e
85	2,20,3,4,40-PeBDE	7.37 ± 0.12ª	7.32	±0.05	0.05	7.1 ^c , 6.72 ^d , 8.06 ^e
99	2,20,4,40,5-PeBDE	7.32 ± 0.14ª	7.27	±0.05	0.05	6.72 ^d
100	2,20,4,40,6-PeBDE	7.24 ± 0.16 ª	7.49	±0.06	-0.25	6.72 ^d
153	2,20,4,40,5,50-HxBDE	7.90 ± 0.14ª	7.58	±0.06	0.32	7.39 ^d
154	2,20,4,40,5,60-HxBDE	7.82 ± 0.16ª	7.89	±0.07	-0.07	7.39 ^d
183	2,20,3,4,40,50,6-HpBDE	8.27 ± 0.26 ^a	8.35	±0.09	-0.08	

 Table A1:
 Experimental and predicted logKow values of selected PBDEs

a Experimental data using slow-stir method, source: Braekevelt et al. 2003.

b Diff.: Difference between observed and predicted $\mathsf{logK}_\mathsf{OW}$ values.

c Source: Palm et al. 2002.

d Mean values of data derived, Source: Hayward et al. 2006.

Location	Sampling year	Ice	BDE-47	BDE-99	BDE-100	BDE-209
lceland to the Fram Strait	2007	Superficial	nd-2.2	nd-1.5	-	-
lceland to the Fram Strait	2007	Deep	nd-2.3	nd-1.6	-	-
Marguerite Bay	2001	Superficial	190±38	200±91	38±14	580±400
Amundsen and Ross Seas	2007	Superficial	32±15	30±9.5	5.5±3.0	2900±3200

Table A2: Concentrations of PBDEs (pg/L) in ice in Polar Regions

nd = not detectable

Source: Lacorte et al. 2009, Dickhut et al. 2012.

Table A3:Concentrations of PBDEs (pg/L) in lake in Polar Regions

Location	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209	PBDE
IHSC	3.4	69	65	15	7.7	8.1	3.2	190	540
SJR	5.6	66	47	13	7.1	5.3	2	310	580
KR	3.2	14	16	4.1	4.3	2	1.4	110	210
GR	6.4	49	38	8.7	6.2	4	2.3	110	290
LFR	2.2	19	19	4.5	5.8	2.8	3.3	350	590

IHSC: Indiana Harbor and Ship Canal; KR: Kalamazoo River; LFR: Lower Fox River; SJR: Saint Joseph River Source: Guo et al. 2017.

Location	Year	BDE-47	BDE-100	BDE-209	ΣPBDEs
Arctic Yellow River Station	2010-2014				1.0 ± 0.3
Antarctic Great Wall Station	2010-2014				0.4
Brisbane River	2014-2015	0.05 ± 0.03 (< 0.003–0.12)	0.01 ± 0.02 (< 0.003–0.05)	4.1 ± 3.1 (< 0.30–11)	4.2 ± 3.1 (0.01–11)
		0.05 ± 0.04 (0.005–0.15)	0.01 ± 0.02 (< 0.003–0.06)	5.0 ± 3.3 (< 0.30–12)	5.2 ± 3.4 (3.4–12)
		0.03 ± 0.02 (0.006–0.08)	0.004 ± 0.001 (< 0.003–0.004)	2.0 ± 0.82 (1.2–3.5)	2.0 ± 0.9 (1.2–3.7)

Table A4: Concentrations of PBDEs (mean and Range, ng/g) in sediment in Polar Regions

Source: Sun et al. 2020, Anim et al. 2017.

Table A5:Concentrations of PBDEs (ng/g) in soil in Polar Regions

Location	Year	BDE-17	BDE-47	BDE-100	BDE-154	BDE-183	RPBDEs
Ny-Ålesund	2013	2.9(0.7-7.5)	4.7(0.9-19)	2.3(0.4-7.2)	4.0(0.7-11)	4.1(0.4-6.7)	42(11-89)

Source: Wang et al. 2015b.

Location	Year	matrix	BDE-28	BDE-47	BDE-99	BDE-100	BDE-154	ΣPBDEs
King George Island	2014- 2015	Chaenocephal us aceratus		6.6	3.6		2.1	3.9
Anvers Island	2002	Juvenile krill		23±3.5	22±3.4	4.5±0.7		
Livingston Island	2000- 2014	Seal	1.9±2.7	20±27	19±32	4.8±8.1		53±7.6
King George Island	2010- 2014	Algae (Palmaria decipiens)						1.5 ± 1.7
King George Island	2010- 2014	limpet (Nacella concinna)						1.0 ± 1.4
King George Island	2010- 2014	Starfish (Diplasterias brucei)						0.34 ± 0.33
King George Island	2010- 2014	Patagonian toothfish (Dissostichus eleginoides)						4.7 ± 10
Ny-Ålesund	2010- 2014	Seaweed (Fucus distichus)						1.0 ± 0.3
Ny-Ålesund	2010- 2014	Amphipod (Themisto libellula)						0.4
Ny-Ålesund	2010- 2014	Barnacle (Balanus Balanus)						10 ± 3
Ny-Ålesund	2010- 2014	Jellyfish (Cyanea sp.)						0.82 ± 0.79
Ny-Ålesund	2010- 2014	Atlantic cod (Gadus morhua)						1.6 ± 0.4

Table A6:	Concentrations of PBDEs (ng/g) in biota in Polar Regions

Source: Sun et al. 2020, Rios et al., Chiuchiolo et al. 2004, Markham et al. 2018.

РАН	Formula	Molar Mass (g/mol)	Sª (mol/m³)	Vp ^a (Pa)	<i>K</i> ow ^a	K oa ^a	<i>K</i> Aw ^a
Naphthalene (Nap)	C ₁₀ H ₈	128.17	-0.09	1.58	3.40	5.19	-1.73
Acenaphthylene (Acy)	C12H8	152.19	-0.57	0.41	3.85	6.46	-2.41
Acenaphthene (Ace)	C12H10	154.21	-0.91	0.24	3.95	6.44	-2.24
Fluorene (Fl)	C13H10	166.22	-1.22	-0.27	4.11	6.85	-2.44
Phenanthrene (Phe)	C14H10	178.23	-1.62	-0.99	4.47	7.64	-2.76
Anthracene (Ant)	C14H10	178.22	-1.94	-1.23	5.57	7.70	-2.69
Pyrene (Pyr)	C16H10	202.25	-2.51	-2.38	5.01	8.86	-3.27
Fluoranthene (Flu)	C ₁₆ H ₁₀	202.25	-2.29	-2.17	4.97	8.81	-3.27
Chrysene (Chr)	C ₁₈ H ₁₂	228.29	-3.44	-3.87	5.67	10.30	-3.82
Benzo[a]anthracene (BaA)	C ₁₈ H ₁₂	228.29	-3.22	-3.42	5.83	10.28	-3.59
Benzo[b]fluoranthene (BbF)	C ₂₀ H ₁₂	252.31	-3.80	-4.98	5.86	11.30	-4.58

Table A7:	Physical and che	mical properties of 16	U.S. EPA priority PAHs
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РАН	Formula	Molar Mass (g/mol)	Sª (mol/m³)	Vp ^a (Pa)	<i>K</i> ow ^a	Koa ^a	K Aw ^a
Benzo[k]fluoranthene (BkF)	$C_{20}H_{12}$	252.31	-3.87	-5.11	5.86	11.36	-4.64
Benzo[a]pyrene (BaP)	C ₂₀ H ₁₂	252.31	-3.98	-5.10	6.05	11.48	-4.51
Benzo[g,h,i]perylene (BghiP)	C22H14	276.33	-4.96	-6.34	6.63	12.55	-4.77
Indeno[1,2,3-c,d]pyrene (IcdP)	C22H12	276.33	-4.88	-6.18	6.57	12.43	-4.70
Dibenzo[a,h]anthracene (DahA)	C22H14	278.35	-3.26 ^e	-7.02 ^f	6.50 ^e	12.59	-

 a logarithmic value at 25°C for S (water solubility), V_p (vapor pressure), $K_{OW},\,K_{OA},\,K_{AW}.$

Source: Hereon

Location	Sampling Year	PAH species; number	∑PAH (dissolved phase)	∑PAH (particulate phase)
Mackenzie Rive, Canadian Arctic	1987	Parent; 12	1.1 ± 0.5ª	120 ± 69
Mackenzie Rive, Canadian Arctic	1993	Parent; 12	12 ± 10	20 ± 8.7
Mackenzie Rive, Canadian Arctic	1994	Parent; 12	8.3 ± 3.1	2.7 ± 0.8
Mackenzie Rive, Canadian Arctic	1994	Parent; 12	13 ± 6.6	260 ± 210
Mackenzie Rive, Canadian Arctic	1987	Alkylated; 12	5.5 ± 3.3	170 ± 100
Mackenzie Rive, Canadian Arctic	1993	Alkylated; 12	44 ± 31	41 ± 21
Mackenzie Rive, Canadian Arctic	1994	Alkylated; 12	79 ± 100	4.3 ± 2.0
Mackenzie Rive, Canadian Arctic	1994	Alkylated; 12	34 ± 12	500 ± 430
Mixing zone, Ob River	1993	Parent + Alkylated; 13		0.41 (0.1-0.7)
Mixing zone, Yenisei River	1993	Parent + Alkylated; 13		0.3 (0.2-0.4)
Riverine zone, Ob River	1993	Parent + Alkylated; 13		1.2 (0.2-2.2)
Riverine zone, Yenisei River	1993	Parent + Alkylated; 13		1.8 (nd-5.1) ^b

Table A8:	Concentrations of P	AHs (ng/L) in rivers in Polar	Regions (mean and range)	ļ

^a Mean ± standard deviation

^b nd = not detected

Source: Yunker et al. 2002, Fernandes and Sicre 1999.

Location	Sampling Year	PAH species; number	ΣΡΑΗ (dissolved phase)	∑PAH (dissolved + particulate phase)
Logne Lake	2012	Parent; 16		340 (150-1200)
Scott Lake	2012	Parent; 16		270 (120-490)
Renard Lake	2012	Parent; 16		76 (20-300)
Antonia Lake	2012	Parent; 16		47 (42-54)
No Worries Lake	2013	Parent + Alkylated; 17	160 ± 13ª, 220 ± 16	
Russian Lake	2013	Parent + Alkylated; 17	360 ± 32	
Lake Reid	2013	Parent + Alkylated; 17	90 ± 7; 350 ± 28	
Lake Sibthorpe	2013	Parent + Alkylated; 17	14 ± 1.1	
Lake Mir	2013	Parent + Alkylated; 17	320 ± 25, 180 ± 15	
Lake Abiskojaure	2008	Parent; 8	12 (Nap) ^b	

 Table A9:
 Concentrations of PAHs (ng/L) in lakes in polar regions (mean and range)

^a Mean ± standard deviation

^b Only Nap in one sample was detected

Source: Lehmann-Konera et al. 2020, Yao et al. 2016, Törneman et al. 2008.

Location	Sampling Year	Soil type	PAH species; number	ΣРАН
South Shetland Islands, Antarctic	2009	Top 1 cm Top 5 cm	Parent + Alkylated	0.30-4.6 0.16-3.5
Svalbard, Arctic	2014	Top 5 cm	Parent; 16	3200 ± 4300ª
Yamal-Nenets autonomous region, Russian Arctic	2017	Top 5-10 cm	Parent; 16	99 (78-130)
James Ross Island, Antarctic	2005	Top 10 cm	Parent; 16	35-170
King George Island and Ardley Island, Antarctic	2018	Top 10 cm	Parent; 16	550–1700
Bulgarian Antarctic Station	2019-2020	Top 10 cm	Parent; 15	170-200
Collins Bay, Antarctic	2015	Top 2 cm	Parent + Alkylated; 36	140 (nd- 52000)⁵
King George Island, Antarctic	2016; 2017	Top 10 cm	Parent; 15	12 (1.9-33)
Deception Island, Antarctic	2009; 2011	Top 5 cm	Parent + Alkylated	20 (4-47)
Barrientos Island, Antarctic	2009; 2011	Top 5 cm	Parent + Alkylated	54 (9-99)
Livingston Island, Antarctic	2009; 2011	Top 5 cm	Parent + Alkylated	54
Penguin Island, Antartic	2009; 2011	Top 5 cm	Parent + Alkylated	66
Fildes Peninsula, Antarctic	2013-2019	Top 5 cm	Parent; 15	155 (21-2200)
Tromsoya, Norway	2013; 2014	Top 0.5–1 cm	Parent + Alkylate; 53	160 (67-210)

Table A10:	Concentrations of PAHs in lakes i	n polar regions	(mean and range)

^a Mean ± standard deviation

^b nd = not detectable

Source: Cabrerizo et al. 2012, Marques et al. 2017, Ji et al. 2019, Klánová et al. 2008, Alekseev and Abakumov 2020, Abakumov et al. 2021, Rodríguez et al. 2018, Deelaman et al. 2021, Cabrerizo et al. 2016, Na et al. 2020a, Casal et al. 2018b.

Compounds	Use	Product period	Estimated total global use/ production (kt)
DDT	insecticide	1950-present	2600
Toxaphene	insecticide	1950-1993	1330
Technical HCHs	insecticide	1948-1997	10000
Chlordane	insecticide	1945-1988	78
Aldrine	insecticide	1950-1992	500
Diedrin	insecticide	1950-1992	34

 Table A11:
 Estimates of the global historical usage or production of some OCPs

Source: Voldner and Li 1995, Li 1999, Barrie et al. 1992, AMAP 2004.

location	sampling time	НСВ	HCHs	Heptachlor epoxide	Chlordane	DDE	Endosulfan	DDD	DDTs	Dieldrin
Alert	2000- 2003			0.57	0.65	0.50	4.3		0.23	0.59
Little Fox Lake	2000- 2003			0.53	0.64	0.47	8.3		0.4	0.31
Kinngait	2000- 2003			0.56	0.7	0.58	3.2		0.36	0.48
Point Barrow	2000- 2003			0.48	0.65	0.69	2.8		0.39	1
Valkarkai	2000- 2003			0.9	1.4	9.9	3.2		12	0.6
Zeppelin	2000- 2003			ND	1.1	0.82	ND		0.41	ND
North Pacific Ocean to the Arctic	2003								1.6- 6.5	
Alert	1993- 2005	29- 70	12-70	0.51-1.2		0.33- 0.71	3.2-6.5	0.033- 0.89	0.17- 0.56	0.49-1.4
Pallas	1996- 2005		12-39			0.47- 0.93		0.01- 0.42	0.078- 0.36	
Storhofdi	1995- 2005	2-14	6.5- 31			0.063- 0.88		0.10- 1.3	0.072- 12	0.10-1.3
Zeppelin	1994- 2006	54- 120	13-35			0.60- 3.3		0.044- 0.20	0.28- 1.9	
Kinngait I	1994	55	92		3.6		5.8		1.6	1.6
Kinngait I	1995	64	80		2.3		5.6		1.4	1.5
Kinngait II	2001	58	30		1.2		2.5		1.2	0.46
Kinngait II	2002	47	30		0.83		3.3		0.63	0.39
Tagish	1993	42	90		2.9		5.8		1.4	0.93
Tagish	1994	55	91		3.2		8.3		2	1.3
Little Fox Lake	2002– 2003	67	52		1		8.3		0.93	0.31
Barrow	2002– 2003	47	22		0.99		2.8		1.2	1
Amderma	1999– 2000	41	32		1		3.7		2.2	0.69

Table A12:The concentrations of OCPs in air (pg/m³) in the Polar Regions

location	sampling time	НСВ	HCHs	Heptachlor epoxide	Chlordane	DDE	Endosulfan	DDD	DDTs	Dieldrin
Amderma	2000– 2001	35	27		0.83		3.5		4.7	0.65
Dunai	1993– 1994	57	55		2.3		3.1		0.98	1.1
Dunai	1994– 1995	64	65		2.1		5.4		1.3	1.3
Valkarkai	2002– 2002	76	71		1.8		3.2		23	0.6

Source: Su et al. 2008; Ding et al. 2009; Hung et al. 2010; AMPAP 2004.

Location	sampling time	sea or lake or river	НСВ	HCHs	Heptachlor	Heptachlor epoxide	Chlordane	DDE	Endosulfan	DDD	DDTs	Dieldrin	Endrin	Aldrin
Japan Sea	2008	sea		2.5	0.48	0.018		0.038	0.092	0.035	0.056	0.025	0.033	0.047
Okhotsk Sea	2008	sea		2.4	0.55	0.016		0.037	0.088	0.031	0.052	0.026	0.036	0.022
Bering Sea	2008	sea		0.92	0.17	0.022		0.040	0.095	0.030	0.055	0.019	0.032	0.24
Chukchi Sea	2008	sea		1.2	0.066	0.021		0.039	0.087	0.026	0.049	0.014	0.027	0.31
Arctic Ocean	2008	sea		2.2	0.20	0.031		0.036	0.097	0.033	0.053	0.018	0.030	0.22
Meteorological station	1999 <i>,</i> 2000	Fog water	27	459										
Meteorological station	2000	Meltwater		1140										
Great Lakes rural	1996-98	lake	68	0.1			9.4				4.5			
Great Lakes urban	1996-98	lake	110	131			151				80			
Alert	1996-98	lake	41	37			2				0.8			
Ny-Ålesund	1996-98	lake	94	56			1.9				2.1			
Stórhöfði	1996-98	lake	9.8	22			0.5				2.6			
Amderma	1999- 2000	lake												
Onega	1990- 1996	lake		0.006				<0.001			<0.001			
Sev. Dvina	1990- 1996	lake		0.002				<0.001			<0.001			

Table A13:The concentrations of OCPs in seawater, lake water and river water (ng/L) in the Polar Regions

Location	sampling time	sea or lake or river	НСВ	HCHs	Heptachlor	Heptachlor epoxide	Chlordane	DDE	Endosulfan	DDD	DDTs	Dieldrin	Endrin	Aldrin
Mezen	1990- 1996	lake		0.008				<0.001			0.004			
Pechora	1990- 1996	lake		0.014				0.001			0.001			
Ob	1990- 1996	lake		0.083				0.002			0.02			
Nadym	1990- 1996	lake		0.094				0.007			0.02			
Pur	1990- 1996	lake		0.18				0.006			0.027			
Taz	1990- 1996	lake		0.17				<0.001			0.038			
Yenisey	1990- 1996	lake		0.025				<0.001			<0.001			
Anabar	1990- 1996	lake		<0.002				<0.001			<0.001			
Olenek	1990- 1996	lake		<0.002				<0.001			<0.001			
Lena	1990- 1996	lake		<0.002				0.001			<0.001			
Kolyma	1990- 1996	lake		<0.004				<0.001			<0.001			
GLI		lake									0.00087	0.001		
OMEE		lake									0.003	0.001		

Location	sampling time	sea or lake or river	нсв	HCHs	Heptachlor	Heptachlor epoxide	Chlordane	DDE	Endosulfan	DDD	DDTs	Dieldrin	Endrin	Aldrin
Lake Ellasjøen	1999- 2000	Fog water	5.2	502										
Lake Ellasjøen	1999- 2000	Fog water	14	830										
Lomonosovfonna	2000	sea	0.003	169			137		85		550	88		
Austfonna	1998	sea	N.D	14			N.D		13		N.D	11		
Summit	2000	sea	8.2	36			105		25		83.6	94		
Lomonosovfonna	2000	sea		57			47		29		187	30		
Austfonna	1998	sea		9.3					8.7			7.5		
Summit	2000	sea	2.4	11			31		7.4		24.5	28		
Meteorological station	2000		2	78										
Ellasjøen	2000		1	32										
Meteorological station	2000	Meltwater		490										
Meteorological station	2000	Meltwater		1800										
Ellasjøen	2000	Meltwater		921										
Ellasjøen	1999	Meltwater		740										
Meteorological station	2000		2	78										
Lake Ellasjøen	2000		1.1	32										

Source: Cai et al. 2010; AMAP 2004; Alexeeva et al. 2001; Johansen et al. 2021.

Compounds	Concentrations in snow of Agassiz Ice Cap, Canada (pg/L)	Total glacial input from Agassiz Ice Cap, Canada (kg)	Flux through the Archipelago (kg/year)
α-HCH	260	200	200000
ү-НСН	110	92	28000
ΣDDT	480	380	160
Chlordane	35	28	96
НСВ	65	52	810
PCBs	3.5	2.8	2700

 Table A14:
 Potential input of selected organochlorines from glacial melt

Source: Macdonald et al. 2005.

Table A15:	Full names, acronyms, chemical abstract system (CAS) numbers of
	perfluoroalkylated substance (PFAS)

Compounds	Abbreviation	CAS No.
Perfluorobutanoic acid	PFBA	375-22-4
Perfluoropentanoic acid	PFPeA	2706-90-3
Perfluorohexanoic acid	PFHxA	307-24-4
Perfluoroheptanoic acid	PFHpA	375-85-9
Perfluorooctanoic acid	PFOA	335-67-1
Perfluorononanoic acid	PFNA	375-95-1
Perfluorodecanoic acid	PFDA	335-76-2
Perfluoroundecanoic acid	PFUnA	2058-94-8
Perfluorododecanoic acid	PFDoA	307-55-1
Perfluorotridecanoic acid	PFTrA	72629-94-8
Perfluorotetradecanoic acid	PFTeA	376-06-7
6:2 Fluorotelomer carboxylic acid	6:2 FTCA	53826-12-3
8:2 Fluorotelomer carboxylic acid	8:2 FTCA	27854-31-5
10:2 Fluorotelomer carboxylic acid	10:2 FTCA	53826-13-4
6:2 Fluorotelomer unsaturated carboxylic acid	6:2 FTUCA	70887-88-6
8:2 Fluorotelomer unsaturated carboxylic acid	8:2 FTUCA	70887-84-2
10:2 Fluorotelomer unsaturated carboxylic acid	10:2 FTUCA	70887-94-4
Perfluorobutane sulfonate	PFBS	375-73-5
Perfluorohexane sulfonate	PFHxS	355-46-4
Perfluorooctane sulfonate	PFOS	1763-23-1
Perfluorodecane sulfonate	PFDS	335-77-3
4:2 Fluorotelomersulfonate	4:2 FTS	757124-72-4
6:2 Fluorotelomersulfonate	6:2 FTS	27619-97-2
8:2 Fluorotelomersulfonate	8:2 FTS	39108-34-4
10:2 Fluorotelomersulfonate	10:2 FTS	120226-60-0
4:2 Fluorotelomer alcohol	4:2 FTOH	2043-47-2
6:2 Fluorotelomer alcohol	6:2 FTOH	647-42-7
8:2 Fluorotelomer alcohol	8:2 FTOH	678-39-7
10:2 Fluorotelomer alcohol	10:2 FTOH	865-86-1
12:2 Fluorotelomer alcohol	12:2 FTOH	39239-77-5
Perfluorosulfonamide	PFOSA	754-91-6
Compounds	Abbreviation	CAS No.
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N-methyl perfluorooctane sulfonamidoethanol	NMeFOSE	24448-09-7
N-ethyl perfluorooctane sulfonamidoethanol	NEtFOSE	1691-99-2
N-methyl Perfluorobutane sulfonamidoethanol	MeFBSE	34454-97-2
N-methyl perfluorooctane sulfonamide	NMeFOSA	31506-32-8
N-ethyl perfluorooctane sulfonamide	NEtFOSA	4151-50-2
N-methyl perfluorobutane sulfonamide	MeFBSA	68298-12-4
6:2 Fluorotelomer acrylate	6:2 FTAC	17527-29-6
8:2 Fluorotelomer acrylate	8:2 FTAC	27905-45-9
3H-perfluoro-3-[(3-methoxypropoxy)propanoic acid]	ADONA	958445-44-8
Hexafluoropropylene oxide dimer acid	HFPO-DA (GenX)	62037-80-3
6:2 chlorinated polyfluorinated ether sulfonate	6:2 CI-PFESA (F-53B)	73606-19-6
Perfluoro-4-ethylcyclohexanesulfonate	PFECHS	335-24-0
1,3-bis-(trifluoromethyl)-5-bromobenzene	BTFBB	328-70-1
6:2 Fluorotelomer phosphate diester	6:2 diPAP	57677-95-9
8:2 Fluorotelomer phosphate diester	8:2 diPAP	678-41-1

Source: Hereon