An Overview on the Legacy and Risks of Polychlorinated Biphenyls (PCBs) and Organochlorinated Pesticides (OCPs) in the Polar Regions

Peng Zhang[a], Jing Wang[a], Andrew Sweetman[b], Linke Ge[a,b,*], Rongguang Xing[a], Hao Ji[a], Jingfeng Yan[a], Qian Xiao[a], Yaqing Cui[a], Hongrui Ma[a], Sisi Xu[c,*]

[a]School of Environmental Science and Engineering, Shaanxi University of Science & Technology, Xi'an, 710021, China

[b]Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK

[c]Resources and Environment Innovation Institute, Shandong Jianzhu University, Jinan 250101, China

Abstract: Polychlorinated Biphenyls (PCBs) and Organochlorinated Pesticides (OCPs) are 'trapped' in a variety of environmental media and can therefore undergo further processing by geochemical cycles. By reviewing a wide range of research studies, we present and discuss the main progresses that affect legacy contaminants, such as migration and transformation processes, biological effects assessment across all Arctic media. PCBs and OCPs demonstrated an overall decreasing concentration trend over time in the Arctic. Ecological risk assessment was undertaken by comparison with two standards, suggesting that there was no ecological risk in either soil or sediment. The concentrations of HCB, ∑HCHs, ∑DDTs, chlordane, mirex, and ∑PCBs increased with trophic levels (TLs), showing a significant linear correlation ($P < 0.001$). The calculated trophic magnification factors (TMFs) values ranged from 0.0004 to 26.63, among which DDTs had the highest value. Future research need to focus its efforts on the exploration of the influence of the long-term fate PCBs and OCPs residues.

* Corresponding author: Dr. Linke Ge (Ge L.)

E-mail: gelinke@sust.edu.cn

Corresponding author: Dr. Sisi Xu (Xu S.)

E-mail: xusisi20@sdjzu.edu.cn

Keywords: Polychlorinated Biphenyls; Organochlorinated Pesticides; Arctic; Multimedia; Biomagnification; Risk

1. Introduction

Twelve organic compounds were classified as Persistent Organic Pollutants (POPs) by the Stockholm Convention in 2001, and are well-known as the Dirty Dozen. The first list of POPs included 9 Organochlorinated Pesticides (OCPs), Polychlorinated Biphenyls (PCBs) and dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) with the characteristics of persistence, toxicity, bioaccumulation, and long-range transport (LRT) potential. The 9 OCPs, included aldrin, chlordane, dichlorodiphenyltrichloroethane (DDT), dieldrin, endrin, heptachlor, hexachlorocyclohexane (HCH), mirex, and toxaphene, which were once extensively used as agricultural and household insecticides. DDT was extensively used to control the spread of vector-born human diseases, like malaria, since the 1940s (Ding et al., 2009; Qiu et al., 2004). OCPs were the dominant pesticides used after the Second World War before they attracted considerable scientific and public concern. After the publication of "Silent Spring" by Rachel Carson in 1962, an increasing audience became aware of the environmental impacts of the widespread use of pesticides. Accordingly, the usage of 9 OCPs were eliminated or reduced gradually across the world. With respect to Polychlorinated Biphenyls (PCBs), they are highly stable industrial chemical products that have been used extensively as industrial fluids, flame retardants, diluents, hydraulic fluids, and dielectric fluids (Zhang et al., 2013).

The carcinogenicity, teratogenicity and mutagenicity of POPs have been well documented already for many years. Toxic effects have been continuously confirmed, such as endocrine disruption, neurotoxicity and so on. Aldrin has been shown to exhibit serious toxicity in animals and human beings, affecting the central nervous system causing symptoms such as seizures (Jhamtani et al., 2018). Numerous studies have showed correlations between technical-grade chlordane (TGC) and reproductive and metabolic disorders (Moreira et al., 2020). Dieldrin, the product of aldrin biotransformation was classified as Group B2, a probable human carcinogen by US EPA (Stern, 2014). Endrin is one of the most toxic organochlorine insecticides with a lethal dose (LD_{50}) of 2.3 mg/kg in mice (Rahman et al., 2018). Heptachlor has been

47 recognized as a human carcinogen with its main mode of toxicity attacking the central nervous system
48 (Purnomo et al., 2013). Toxaphene, the most widely applied pesticide in the world, was used as an
49 alternative of DDT in the early 1970s. It has been discovered to be highly toxic to fish and rodents and is
50 probably also a human carcinogen (Veyrand et al., 2008). PCBs have been acknowledged to exhibit
51 neurotoxic and endocrine disrupting effects on birds and mammals in polar regions. More specifically,
52 chiral PCB congeners, particularly those with 2,3, and 6 substitution patterns on one phenyl ring, have been
53 associated with neurodevelopmental toxicity in animals and humans, revealing their effects on
54 neurotransmitter functions in the central nervous system and altering the cellular processes concerning
55 calcium signaling (Wu et al., 2014). OH-PCBs have also demonstrated several mechanisms of influence,
56 for example, disrupting cellular calcium homeostasis by mechanisms including RyRs (Niknam et al., 2013)
57 or leading to thyroid dysfunction (Kodavanti and Curras-Collazo, 2010; Meerts et al., 2002; Wu et al.,
58 2014).

59 Due to their semi-volatility, persistence and long-range transport potential, OCPs and PCBs are able
60 to transfer to pristine polar areas. An abundance of studies, reports issued by monitoring programs and book
61 chapters, have demonstrated that PCBs and OCPs are ubiquitous in polar areas, including the atmosphere,
62 water, soil, sediments and biota ranging from zooplankton to top predators. Several regional and
63 international monitoring programs have been undertaken in order to comprehensively explore the status of
64 POPs in the polar regions, such as the Arctic Monitoring and Assessment Programme (AMAP), United
65 Nations Economic Commission for Europe, European Monitoring and Evaluation Programme (UNECE-
66 EMEP) and Global Atmospheric Passive Sampling (GAPS) Network. Polar areas are considered as
67 “primary sink” of POPs, although they may become a “secondary source” due to global warming.
68 Specifically, elevated temperatures in polar areas accelerates snow and glacier melting facilitating
69 volatilization from water. In this present study, progress on the occurrence, environmental behavior and
70 ecological risks of PCBs and OCPs in the Arctic have been reviewed. The objectives of this present study
71 were to: 1) present the temporal and spatial distribution of PCBs and OCPs in various environmental

matrices and biota, 2) assess their sources and transport path to polar areas and 3) evaluate the ecological risks and biological effects from exposure.

2. Data collection

Based on various combinations of keywords, such as “organochlorine pesticides”, “polychlorinated biphenyls”, “Arctic”, “bioaccumulation” or “biological impact”, a total of 1241 studies for OCPs and 680 studies for PCBs were retrieved from the “web of science”, including relevant cross-references. These studies provided decades of concentration data for OCPs and PCBs, reporting their environmental behavior, and assessing their biomagnification through food webs. The literature sources were exported and analyzed using VOS-viewer software (version 1.6.19) to produce a cluster view. As Fig. 1 shows, cluster 1 mainly targeted the pollution of pesticides (such as Current Used Pesticides, CUPs) and OCPs. Cluster 2 mostly related with investigations within the Arctic; cluster 3, cluster 4 and cluster 5 were joint studies with PCBs and OCPs and their influencing factors. Cluster 6 and cluster 7 primarily exhibited research with correlations between specific pollutants and OCPs and PCBs. Cluster 8 mainly focused on the biological effects caused by pollutant exposure, such as biological amplification. According to data related to polar OCPs and PCBs, measured concentrations in the atmosphere, water, soil, sediment, plants and animals were compared, mapped and analyzed in order to provide insights into the protection of the Arctic ecological environment and the promotion of human health.

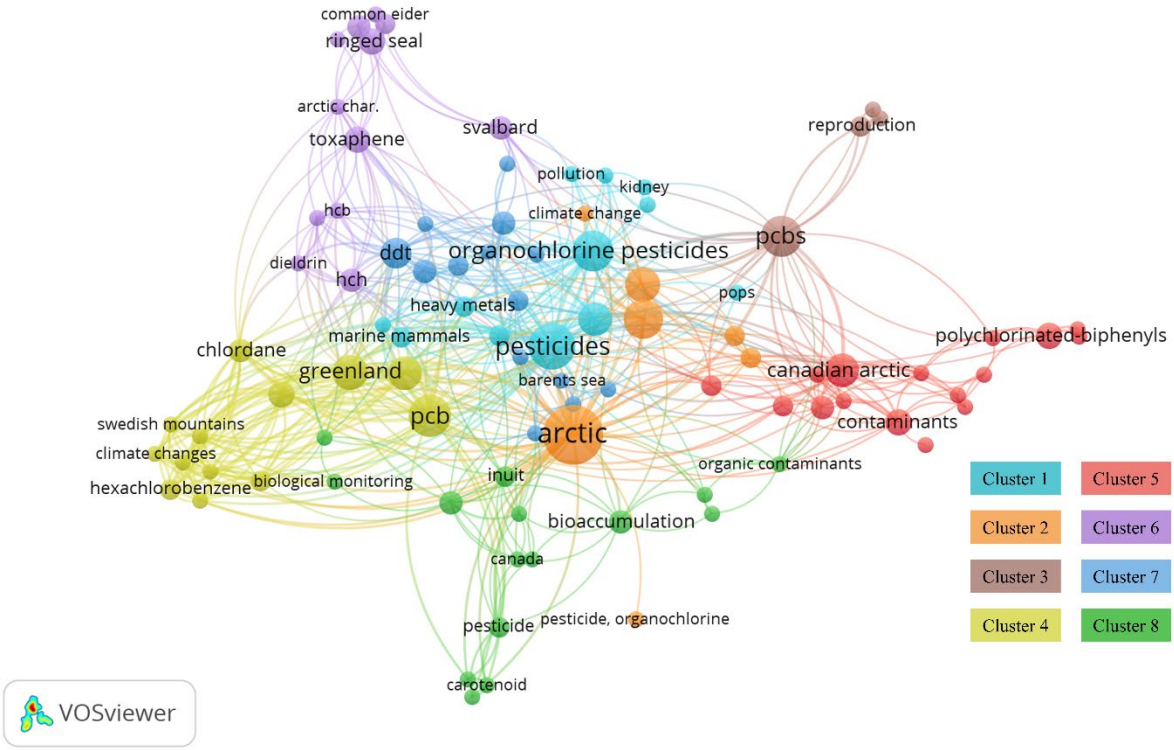


Fig. 1. Cluster analysis of literature data related to PCBs and OCPs.

3. Sampling approaches and analysis methods in polar areas

The Arctic locations included in the studies are shown in Fig. S6, including the Arctic Ocean and parts of Canada, Russia, Norway, Sweden, Finland and Iceland, and the United States. Both passive and active sampling approaches have been frequently used as atmospheric POPs monitoring tools in polar areas. Polyurethane foam disk (PUF) is a common absorption phase and used in both active and passive samplers (Bidleman et al., 2017; Cabrerizo et al., 2017; Li et al., 2012). The GAPS network uses PUF-disks and includes sampling stations at Barrow, Ny-Ålesund, Stórhöfði (Storhofdi) and Little Fox Lake (Vorkamp et al., 2019). Plants, including moss, lichens, dwarf birch (*Betula glandulosa*), have also been considered as promising passive samplers for POPs monitoring (Luttmer et al., 2013). With respect to water samples, large volumes of water are extracted using solid phase (SPE) cartridges whilst liquid-liquid extraction was seldom applied because the low concentrations in water required several litres of water to reach the Limits of Detection (LODs). More recentl a novel passive sampler, triolein-embedded cellulose acetate membranes

(TECAM) has been deployed in the coastal area of Fildes Peninsula, Antarctica, sampling 66 organic contaminants in sea water (Gao et al., 2018). (Gao et al., 2019) also conducted passive sampling of organic pollutants during an Arctic cruise using a self-developed device in combination with TECAM. TECAM may also be applicable to passive sampling of PCBs and OCPs in Arctic seawater.

For solids, including soil, sediment, vegetation, birds, fish, mammals, as well as their feces, samples underwent a stringent process consisting of pretreatment, clean-up and instrument analysis according to existing analysis guidelines or established methods. Soxhlet extraction, ultrasonic extraction and accelerated solvent extraction were the three preferred extraction methods. Gel permeation chromatography (GPC), multi-silica gel column, SPE or combination methods were usually used to eliminate proteins, pigments and other co-extracted interfering substances in samples.

4. Sources and long-range transport paths to polar areas

PCBs and OCPs have been introduced into pristine polar areas through several pathways, involving the air mass movement (Hao et al., 2019; Montone et al., 2003; Ubl et al., 2012; Vecchiato et al., 2015; Wania, 2003), ocean currents (Kallenborn et al., 2013; Ma et al., 2015), 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 so on), revolatilization from oceans and melting glaciers on account of 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).

4.1. Atmospheric sources and long-range transport

It is well established that PCBs and OCPs have been introduced into the pristine Arctic from their production and usage areas via long-range transport. The “grasshopper effect” of atmospheric long-range transport via repeated volatilization/condensation steps has played a dominant role in the presence of legacy of POPs in the Arctic (Aslam et al., 2019; Gouin et al., 2004). The existence of OCPs in polar areas has been attributed to cold condensation and global fractionation during long-range atmospheric transport (LRAT) (Wania, 2003). Three southern air mass routes have contributed to transport in winter: the

Norwegian Sea (40%), Eastern Europe/Siberia (15%), and Bering Sea (25%), respectively, which accounts for about 80% of the annual north-south atmospheric transport (Iversen, 1996). Thus, atmospheric transport was the main pathway in winter. On the contrary, summer air pressure is comparatively higher in the polar regions than in the middle and high latitudes, hindering the incursion of Arctic of air masses (Macdonald et al., 2005).

The presence of PCBs in the Arctic have been attributed mostly to Europe, Asia and Canada (Macdonald et al., 2000; Malanichev et al., 2004; Wu et al., 2011). Some models have been used to predict the propagation distance of PCBs. For instance, the estimated Characteristic Travel Distance (CTD) of PCBs has been reported to be in the range of 578 km (PCB194) ~ 7411 km (PCB52) by the TaPL3 model, and 1484 km (PCB194) to 26908 km (PCB153) by the ELPOS model (Wania, 2003; Wania and Dugani, 2003). The octanol-water partition coefficient (K_{OW}) and octanol-air partition coefficient (K_{OA}) values of PCBs were considered as two key influencing factors for their migration and transformation. The Globe-POP model inferred that PCBs were transferred from low latitudes to the Arctic via multiple jumps of repeated cycles of deposition and re-evaporation, suggesting that the chemicals with log K_{OW} values of 5 ~ 8 (log K_{OA} ranged from 6.5 to 10) undergo air-surface exchange. Nevertheless, less volatile chemicals (log $K_{OA} > 9.5$) were unlikely to take "multiple jumps" as they did not volatilize effectively after being deposited into water or soil, and so they had to arrive in the Arctic with a single atmospheric LRT event (Wania, 2003).

4.2. Currents, rivers, ice and snow input

Ocean currents and riverine input have been important to the arrival of OCPs in the Arctic Ocean. For decades, sediment release, sewage discharge and soil leachate infiltration have been the main ways for OCPs to flow into rivers. Ocean currents and rivers from the source areas served as significant pathways (Ma et al., 2015). For example, the rivers flowing from Russia were regarded as important to organochlorine compounds transport via estuaries and regional seas, particularly those of close to the Arctic basin (Alexeeva et al., 2001; Zhulidov et al., 2000). The main input route of α-HCH into the Arctic Ocean was

atmospheric LRT from 1945 to 1990, while it has been substituted by ocean currents since the early 1990s (Li and Macdonald, 2005). With the banning of technical HCH usage during the 1980s, the transition of the Arctic Ocean from a sink to a source of HCH in the atmosphere occurred due to the atmospheric α-HCH concentrations decline. It has been well documented that residual HCHs in the cold, ice-covered surface waters of the western Arctic Ocean continue to supply HCH to out-flowing waters in the Canadian Archipelago (Pućko et al., 2013). Moreover, the re-volatilization and release of OCPs from glaciers, attributed to the recent melting of ice and snow on account of global climate change, are considered as potential important secondary sources (Bogdal et al., 2009; Khairy et al., 2016; Macdonald et al., 2005).

4.3. Sources apportion associating with their usage

The ratio of OCP isomers, also known as their fingerprint, are regularly utilized to determine the sources and status of their industrial production. There are two sources of HCHs in the environment, technical HCHs and Lindane. The ratio of α-HCH/γ-HCH is an indicator of historical usage, recent technical HCHs or Lindane input. The discrepancy in the ratio of α-/γ-HCH isomers suggests a distinct possible source. For example, there was a significant difference in the ratio of α-/γ-HCH isomers between the Arctic (10.2) and Antarctic sites (2.5) (Nash et al., 2017). The reason for this phenomenon can be explained by the fact that the northern temperate and cold zones were the main sources of α-HCH, while the recent usage of Lindane in the southern hemisphere led to a higher proportion of γ-HCH in the waters of the Antarctic Ocean (He, 2013).

The comparative proportions of the parent compound DDT and its metabolites DDD and DDE were indicators for investigating potential sources of DDT residues in the environment. The relative abundance of parent and metabolite has been used to differentiate fresh ($\sum$DDT/$\sum$DDE > 1) and aged ($\sum$DDT/$\sum$DDE < 1) composition. The increasing content of ΣDDTs (DDT and its isomers) suggested fresh utilization of DDT in the corresponding regions (AMAP, 2004). To date, technical DDT was the predominant source. DDT in the Arctic was not as fresh as in the Far East by comparison. For dicofol (related to DDT), the

177 contribution was estimated to be low. “New” *o,p*'-DDT has been observed in the North Pacific and Arctic,
178 indicating that DDTs were more likely to derive from dicofol type DDT (Ding et al., 2009).

179 Technical chlordane, consisting of a mixture of *trans*-chlordane (TC) and *cis*-chlordane (CC), is the
180 present primary source of residual insecticide in the ecological environment. CC has been found to degrade
181 faster than TC, resulting in the gradual decline of the concentrations ratios. Therefore, the isomer ratio could
182 be used to assess historical legacy or recent input. Generally the CC/TC proportion in the range of 1.2 ~ 1.3
183 demonstrates that fresh technology chlordane products enter the environment (Lin et al., 2012).

184 *4.4. Animal transport*

185 Some animals such as sea birds, whales, pinnipeds, salmon and cod, etc., are migrate long distances,
186 travelling through many human-intensive industrial and agricultural areas, countries, eventually reaching
187 polar regions. To date, some studies have speculated that animal migration was another confirmed route for
188 OCPs to enter polar regions. Organic contaminant concentrations were found to be twice as concentrated
189 in salmon spawning lakes as in salmon-free lakes, with the composition of compounds found in salmon
190 being similar. Bio-transport was suggested to have a greater influence to PCBs and DDT loadings in lake
191 biota than atmospheric inputs in Alaskan river systems (Ewald et al., 1998). Seabirds have also been
192 reported to be involved in bio-transport of certain OCPs into the Arctic, resulting in OCPs in bird habitats
193 being elevated several fold higher than corresponding background concentrations (Blais et al., 2005; Choy
194 et al., 2010; Evenset et al., 2004; Michelutti et al., 2009; Wania, 2003).

195 **5. Transformation processes**

196 Several studies have shown that PCBs changed their chiral characteristics in the environment after
197 long-distance transport through air, oceans and lakes, although the mechanisms are not clearly understood.
198 For example, changes in the enantiomer fraction (*EF*) of PCB136 have been observed in equatorial Indian
199 Ocean (Huang et al., 2013) and Arctic regions (Bidleman et al., 2012). Fresh PCBs which were released
200 into air via air-water or air-soil exchange played an important role in determining the PCB *EF* values.

It has been demonstrated that OH radicals in air oxidize PCBs in the gas phase (Mandalakis et al., 2003). A five-year atmospheric monitoring study demonstrated that some of the lighters chlorinated PCBs showed a significant decrease in the Arctic atmosphere due to the degradation by OH radicals. Whereas, for heavier congeners, precipitation or removal processes such as particle deposition and snow removal may be more significant (Hung et al., 2001).

After PCBs entering an organism, they can be transformed into water-soluble compounds through phase I and II biotransformation enzymes. Phase I cytochrome P450 enzymes, for example, play an important role in metabolizing and eliminating mono-ortho or *non-ortho* chlorine congeners of PCBs. CYPs catalyze direct insertion of an OH-group into a PCB molecule to form arene-oxide intermediates through which OH-PCBs is formed. In addition, one of the phase II enzymes, glutathione S-transferase (GST), formed methyl sulfones ($MeSO_2$-PCBs) via hydrolytic coupling of glutathione (GSH) to peptidases (Helgason et al., 2010). OH-PCBs and $MeSO_2$-PCBs, two metabolites, have been found in Arctic seabirds, including glaucous gulls, northern fulmar chicks and even in arctic foxes (Helgason et al., 2010; Nost et al., 2012; Routti et al., 2016; Verreault et al., 2007; Wu et al., 2014). The biotransformation of PCBs in biota was found to be highly species-dependent. Generally, invertebrates have a lower capacity to metabolize PCBs compared to fish, birds or mammals.

OCPs can also be converted under some circumstances in the natural environment. DDT, can be transformed to DDE and DDD through oxidation and reduction reactions, respectively. The diverse physical and chemical properties of each HCH isomer have led to differences in their stability in the marine and atmospheric environments. γ-HCH is susceptible to degradation to α-HCH in the via microbial metabolism and photochemical reactions (Walker et al., 1999), while α-HCH can be converted to β-HCH which has a more stable symmetric structure (Ya et al., 2017). In studies of heptachlor and its metabolites, it has been proposed that heptachlor metabolites would undergo the initial metabolic transformation processes of epoxidation, dechlorination and hydrolysis (Purnomo et al., 2013). The parent chlordane compounds were

also transformed under environmental conditions, into oxychlordane and heptachlor *exo*-epoxide, two stable degradation products (Su et al., 2008).

6. Legacies and temporal tendencies in multimedia of polar areas

6.1. Legacies and temporal tendencies of PCB in the multimedia of the polar areas

Data from previously published studies on the occurrence of PCBs in atmospheric, aquatic and solid samples along with biota are illustrated in Fig. 2.

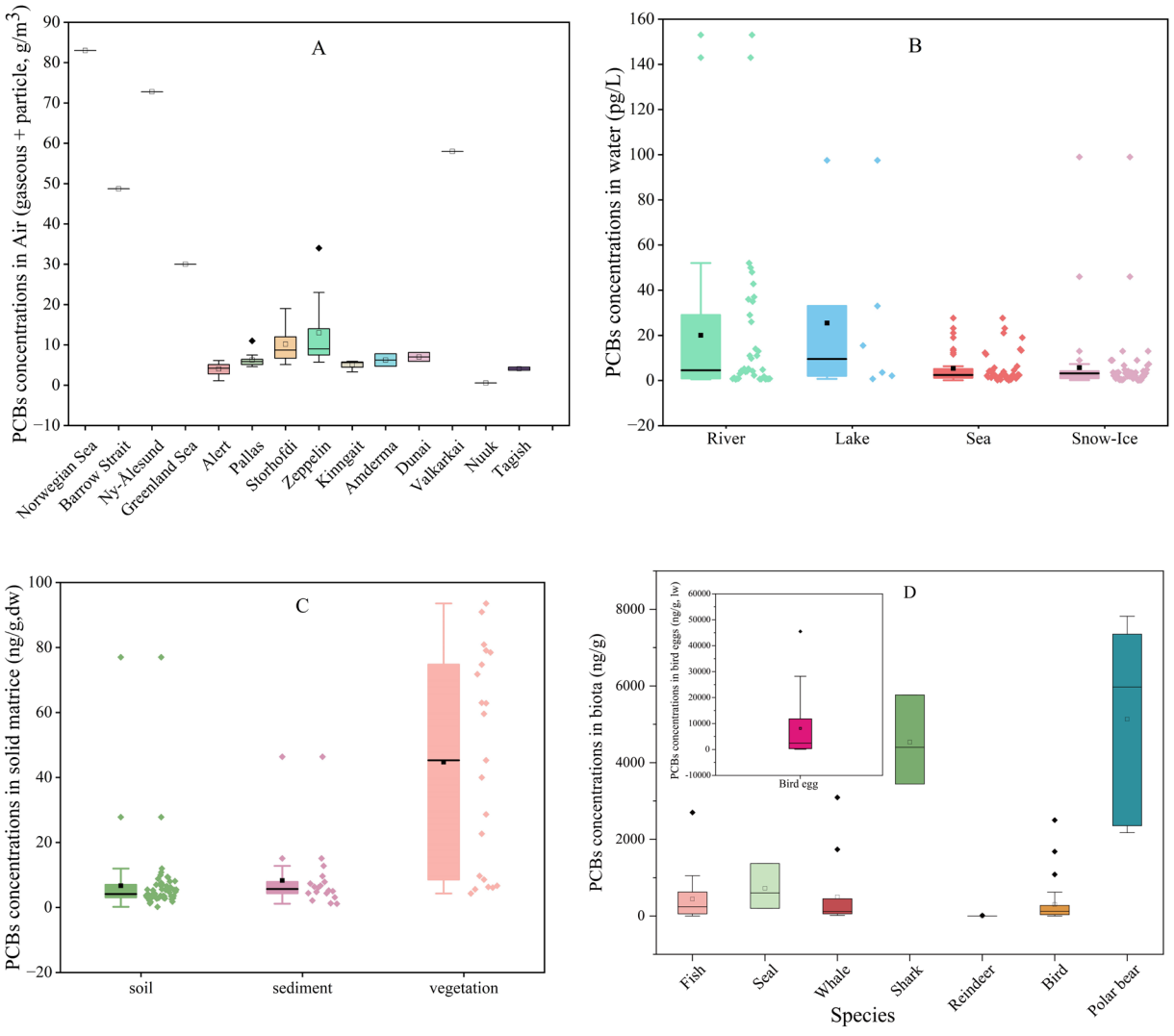


Fig. 2. The PCBs concentrations in air (pg/m^3), water (pg/L), solid matrices (pg/g dw) and biota (ng/g) in the Arctic. The original data was retrieved from a range of studies, whose references were listed in Tables S1 ~ S7 of Supporting Information.

236 *6.1.1. Atmosphere*

237 Atmospheric PCBs concentrations in Arctic air have been reported to be in the range of < LOD to
238 several 10's of pg/m^3. The highest concentrations were found in the Norwegian Sea, followed by Ny-
239 Ålesund, which reached 83 and 72.8 pg/m^3, respectively. The lowest concentration was reported in Nuuk
240 at 0.56 pg/m^3 (Fig. 2A; Table S1). Long-term monitoring results demonstrated a general downward trend
241 for most legacy POPs in Arctic air (Hung et al., 2010; Hung et al., 2016). In the west coast of Sweden and
242 the subarctic region of northern Finland, the temporal tendencies of declining atmospheric concentrations
243 of PCBs, HCHs, chlordane and DDTs were recognized at the rates of 2 ~ 4%, 6 ~ 7%, 3 ~ 4% and 2 ~ 5%
244 per year, respectively (Bargagli, 2008). Monitoring data have consistently demonstrated that the levels of
245 most PCBs in the Arctic atmosphere have declined differently over the past few decades.

246 *6.1.2. Water*

247 PCB concentrations in Arctic sea water and lake water have been found to be comparable, ranging
248 from < LOD to several 10's of pg/L (Fig. 2B; Tables S2 - S3). Nevertheless, the concentration of PCBs in
249 ice and snow varied significantly by 3 orders of magnitude. In general, PCBs concentrations in sea water
250 and lake water were much lower than in snow and ice. Studies have reported that the concentrations of
251 PCBs in the West and East Lakes of Melville Island, as well as in seawater, have increased sharply during
252 the non-glacial period, possibly due to the melting of the ice layer and the input of river runoff (Cabrerizo
253 et al., 2019). Consequently, the concentrations trends suggest that snow/ice melting is serving as a
254 secondary source of PCBs in Arctic Ocean.

255 *6.1.3. Solid media*

256 The residues of PCBs in solid media of the Arctic ranged from < LOD to 10's of ng/g, with higher
257 accumulations in plants (Fig. 2C; Tables S4 ~ S6). Mean concentrations of PCBs in lichens and mosses
258 were usually several times higher than those in abiotic substrates. Results from a number of studies have
259 suggested that total PCBs concentrations ($\sum_{29}$PCBs) in soil, sediment, plant, bird guano and reindeer feces
260 in the Ny-Ålesund region of the Arctic were in the range of 2.76 ~ 10.8 ng/g, 3.09 ~ 8.32 ng/g, 22.5 ~ 56.3

ng/g, 35.4 ~ 51.4 ng/g, and 31.8 ~ 39.6 ng/g dry weight (dw), respectively (Zhang et al., 2014). Average ΣPCBs concentrations in vegetation (6.90 ± 0.81 ng/g dw) were obviously higher than that in soils (3.70 ± 0.36 ng/g dw) (Aslam et al., 2019). PCBs were negatively correlated with elements derived from local bedrock, suggesting that their levels may be impacted by atmospheric deposition.

6.1.4. Biota

PCBs bioaccumulate in Arctic organisms to significantly different extents (Fig. 2D; Table S7). Concentrations of PCBs in birds' tissues varied from < LOD to several thousand ng/g, but reached up to tens of thousands ng/g in eggs. The concentrations of PCBs were all significantly higher than those detected in Ivory Gull eggs collected from the Canadian Arctic in 1976, 1987, and 2004, with concentrations approximately 4 ~ 8 times higher than those reported in 2004 (Su et al., 2006). The concentrations of PCBs in polar bears exceeded 7,000 ng/g with higher concentrations in adipose tissue than in the liver. The discrepancy was possibly caused by the differences in biochemical processes and liver biotransformation (Gebbink et al., 2008). Bioaccumulation of PCBs in the adipose tissue of polar bears may be attributed to the exposure of commercial PCBs and their mixtures (Verreault et al., 2005). The average concentration of PCBs in the plasma of Greenland sharks from Kongsfjorden (Svalbard) reached 5776 ng/g (Neerland et al., 2019), and the average PCB concentration analyzed in the muscle and liver of Greenland sharks from Iceland was 4100 and 4400 ng/g, respectively. This difference could be related to the parts of the body sampled, feeding habits, and nutritional status (Strid et al., 2007). Three compounds of *non-ortho*-, *mono-ortho*- and *di-ortho*-substituted PCBs congeners (NO-CB, MO-CB and DO-CB) were detected in the lipid of ringed seals in Holman (Canadian Arctic) during the 1980s and 1990s without obvious temporal variation in their total concentrations (Addison et al., 2005). However, PCB concentrations in Baltic Sea fish measured along the Finnish coast of the Baltic Sea have decreased by approximately 80% from 1978 to 2009 (Airaksinen et al., 2014). PCBs concentrations in the liver of arctic foxes (*Vulpes lagopus*) have declined at a rate of 4 ~ 11% per year. It was probably due to changes in the diet associated with climate change that influenced concentrations in the arctic foxes from Svalbard (Andersen et al., 2015).

Zooplankton represent an important link between the base of the food web and higher trophic levels (TLs) (Lomartire et al., 2021). A study reported that the mean concentrations of PCBs in herbivorous zooplankton from Isfjorden, Svalbard showed seasonal variation, with concentrations of 4.43 ng/g in May and 1.6 ng/g in August (McGovern et al., 2022).

PCB concentrations in biota (particularly in seabirds and mammals) were approximately 3 ~ 4 times higher than those in environmental media, suggesting that accumulation and magnification of PCBs occurred in organisms. The Baltic food web model predictions revealed that the equilibrium lipid concentrations of all PCB congeners increased with position, with the highest concentrations predicted for cod and salmon (Nfon and Cousins, 2007). The bioaccumulation factors (BAF, $C_{organism}/C_w$) of 4 PCB congeners in the food webs of the Barents Sea (Arctic zone, 77 °N ~ 82 °N) and Baltic Sea (temperate zone sea, 54 °N ~ 62 °N) were compared, both of which included zooplankton, fish, and seal. The BAF_{arctic}: $BAF_{temperature}$ ratios of the four PCB homologues in zooplankton and fish were all greater than 1, ranging from 6.4 to 13.8 and 2.9 to 5.0, respectively, while the ratios in seals were less than 1.0 (Sobek et al., 2010). Both the above model and the investigation revealed that temperature had an important impact on the bioaccumulation of PCBs via food web (Cabrerizo et al., 2018b; Nfon and Cousins, 2007; Sobek et al., 2010). Trophic magnification factors (TMFs) are important parameters that characterize the degree of biological amplification. Some researchers have found that legacy PCBs and DDTs were significantly amplified via the TLs, with TMFs values above 1.0 (Kim et al., 2021).

In summary, there is strong evidence of intentionally manufactured PCBs remain in the Arctic environment, but the recent detection of unintentionally produced PCBs (UP-PCBs), including PCB5 and PCB11, have become a growing concern as well (Bartlett et al., 2019). Surface snow gathered in the Lomonosovfonna region of Svalbard in 2009 ~ 2010 and 2013 ~ 2014 had a high content of PCB11, accounting for 4% of the entire PCBs (Garmash et al., 2013). PCB11 and other UP-PCBs have not been fully explored in Arctic yet. Even though these UP-PCBs were not as high in concentration as some residual

PCBs, the toxicity of the parent and metabolite remains vague, which may lead to unknown negative ecological impacts.

6.2. Legacies and temporal tendencies of OCPs in multimedia of the polar areas

6.2.1. Atmosphere

Air concentrations of OCPs have been summarized at seventeen Arctic stations during 1993 to 2017, including Alert, Kinngait, Tagish, Little Fox Lake, and Cornwallis Island in Canada; Pallas in Finland; Storhofdi in Iceland; Zeppelin, Meteorological station, Tromsø, and Andøya in Norway; Point Barrow in the USA; Amderma, Dunai, Valkarkai, and Tiksi in Russia; and Villum Research Station (VRS) in Greenland see Fig. 3 and Table S8. Concentrations of OCPs monitored at each station varied considerably, with OCPs average concentrations at the stations of Alert (Canada), Zeppelin (Norway), Kinngaint I and II (Canada), Tromsø (Norway) and VRS (Greenland) were notably higher than other stations. With respect to the temporal variation, a generally decreasing trend were observed at most of the stations. HCHs and HCB took the top two places alternatively among the total OCPs in Canada and Norway, implying the different usage strategies in two countries. Air concentrations of HCB reported at all the stations except Storhofdi were high, with mean air concentrations ranging from 29 to 120 pg/m^3 from 1993 to 2006. After 2015, all stations exhibited relatively low air concentrations ranging from an average of 10 to 78 pg/m^3 except for Tromsø, which presented the highest air concentrations of 118 pg/m^3 in 2013 ~ 2014. Because the sampling campaigns were executed mainly in warmer months, the atmospheric concentrations demonstrated obvious seasonal variation characteristic, that is, the higher concentrations were determined during the warmer months, while the lower concentrations were observed during the colder months. Furthermore, gas-particle partition was also affected by temperature leading to higher gas phase concentrations at higher temperature (Casal et al., 2018).

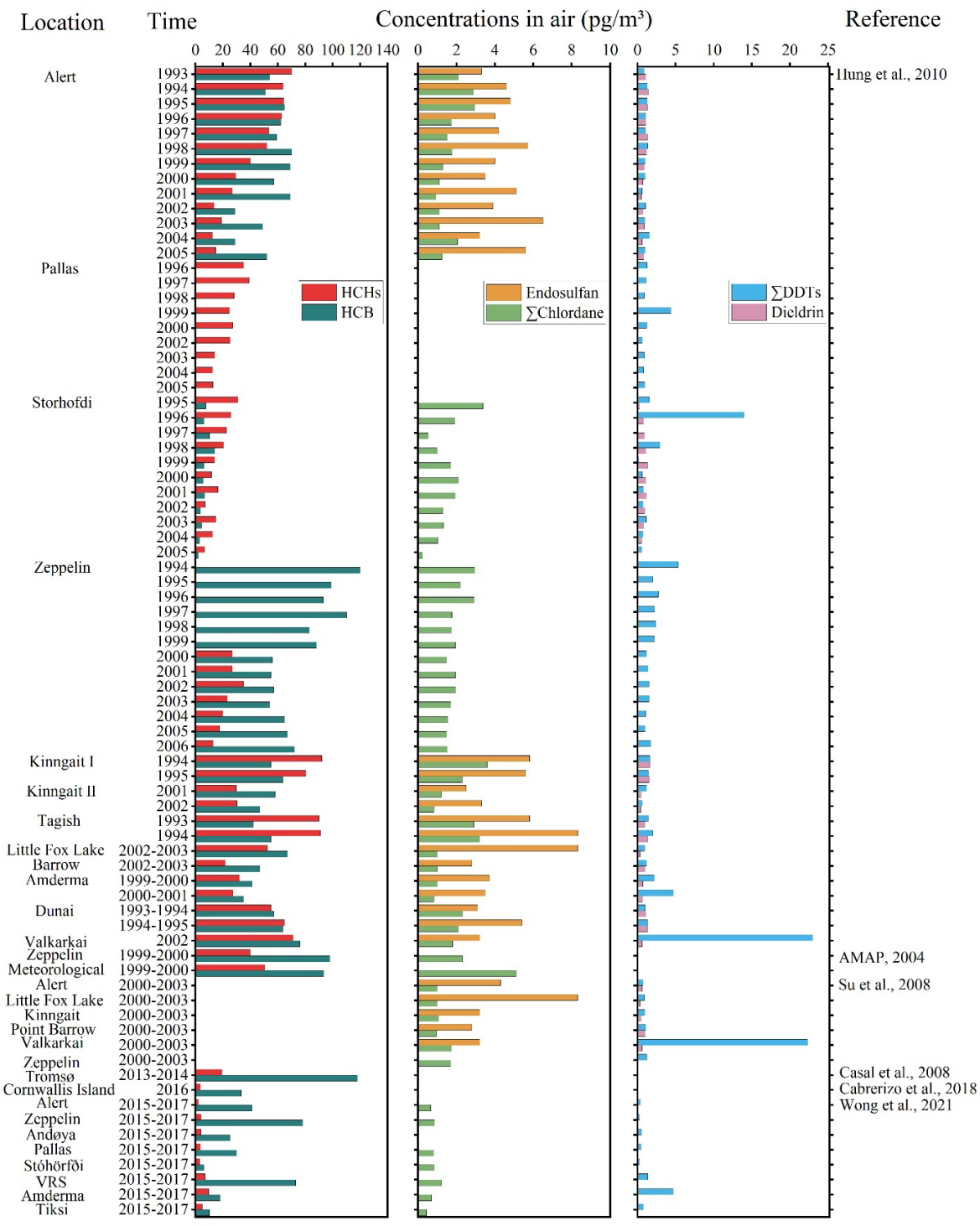
Air concentrations of HCHs measured during the period 1993 ~ 2006 ranged from 6.1 to 92 pg/m^3 and had been decreasing annually. After 2000, air concentrations dropped to 2.1 ~ 34.9 pg/m^3 at all stations, except for Little Fox Lake and Valkarkai (Su et al., 2006). The higher air concentrations detected at Little

Fox Lake were attributed to secondary emissions of technical HCH that had previously deposited in the soil. Although the use of technical HCH resulted in an increase of HCH concentration, higher temperatures were conducive to HCH volatilization (Su et al., 2006). Higher concentrations may also be attributed to the increasing volatilization previously deposited in the ocean, sea ice cover reduction and the continuous usage of some pesticides containing HCH (Hung et al., 2010). After 2015, the concentrations of HCHs were less than 10 pg/m^3. The decrease in air concentrations of α-HCH and γ-HCH across Arctic stations was probably on account of the reducing global emission of technical HCHs and lindane (Wong et al., 2021).

Technical chlordane was primarily composed of *trans*-chlordane (TC), *cis*-chlordane (CC), *trans*-nonachlor (TN), *cis*-nonachlor (CN), as well as traces of heptachlor (HEPT) and some other contaminants (Hung et al., 2010). The concentration of ∑chlordane at all stations were reported as the sum of four chlordane-related compounds, including TC, CC, TN, CN, with the exception of Pallas. The ∑chlordane in the air from 1993 to 2006 ranged from 0.83 to 3.60 pg/m^3, while all stations exhibited relatively lower air concentrations ranging from 0.427 to 1.198 pg/m^3 after 2015. Su et al. (2008) demonstrated that the seasonal variation of chlordane-related compounds was unremarkable in that the monthly mean values of TC, CC, and TN varied within a factor of 4 at five stations (excluding Valkarkai). Seasonality of chlordane-related substances might be ascribed to other factors, involving inputs by LRT, degradation, temperature and atmospheric mixture heights variations as well.

Endosulfan was a broadly applied CUP on plants, such as cotton, tomatoes, and apples. Endosulfan I was reported with the range of 2.5 to 8.3 pg/m^3 at Alert and all satellite stations, including Kinngait, Tagish, Little Fox Lake, *etc*. The highest concentrations reported at Tagish in 1994 and Little Fox Lake in 2000 ~ 2003 were both 8.3 pg/m^3, and the same concentration was measured again at Little Fox Lake in 2000 ~ 2003 by another study. Since the two sites were located in the western lower reaches of the North American Arctic, they may be affected by air masses from Asia and western North America, where endosulfan was frequently used (Hung et al., 2010).

359 Concentrations of DDTs were relatively lower in the Arctic atmosphere, when compared with HCB
360 and HCHs. Air concentrations of *p,p*'- and *o,p*'-DDD were predominantly lower than Method Detection
361 Limit (MDLs) in many studies. The concentration of DDTs in the atmosphere were less than 2.97 pg/m^3 at
362 almost all stations except Valkarkai in 2000 ~ 2003 (22.28 ~ 23 pg/m^3), Storhofdi in 1996 (13.98 pg/m^3).
363 Valkarkai is a distant coastal meteorological station, with probable sources of DDT-related compounds at
364 related to weathered sources (Su et al., 2008). This explanation was also demonstrated in Hung's study
365 (Hung et al., 2010) who reported air concentrations of DDTs in Storhofdi at 13.98 pg/m^3 in 1996.
366 Dieldrin was found at low concentrations, quite close to detection limits in Arctic air, except on a few
367 occasions. Dieldrin concentrations in air ranged from MDL ~ 1.6 pg/m^3, as shown in Fig. 3. On one hand,
368 atmospheric concentrations of dieldrin in the Arctic in 2000 ~ 2003 dropped approximately by half
369 compared with the results in the 1990s because the half-life of dieldrin was about 13 years in the Arctic
370 atmosphere (Hung et al., 2005; Su et al., 2008). On the other hand, some studies have shown that significant
371 negative temperature-dependent relationship, suggesting that the volatile emissions might lead to the high
372 summer air concentrations (Su et al., 2008).



373

374 **Fig. 3.** The concentrations of OCPs in gaseous phase and particle phase of air (pg/m^3) in the Arctic. The

375 original data was retrieved from many researches, whose references were listed in Table S8 of Supporting

376 Information.

6.2.2. Water

HCB was detected in fresh water and sea water with average concentrations in the range of 1.41 to 6.4 pg/L (Fig. S1, Table S9 - S10), while HCB was measured at Svalbard for the first time and dominated the aqueous phase, ranging from 14 to 66 pg/L with maximum concentrations in Ebbaelva in 2018 (Johansen et al., 2021). The differences in concentrations indicated that they were the result of a combination of source distribution, biochemical processes, and environmental conditions.

The concentrations of HCHs in fresh water and sea water of the Arctic were in the range of 22.09 to 3900 pg/L. The general pattern of OCPs in seawater was dominated by HCH (Cai et al., 2010). The concentrations of HCHs at Chukchi Sea and Bering Sea in 1999 were only about one seventh of the data reported by Chernyak et al. (1995) in 1988, due to the prohibition of HCHs in the majority of northern hemisphere nations from the early 1970s (Yao et al., 2002). The total concentration of the 17 classic OCPs increased from the Sea of Japan to the Arctic Ocean via the Okhotsk Sea and Bering Sea in that order. The different latitudinal trends of α-HCH and γ-HCH were discussed in combination with their differences in thermodynamic properties, which might lead to slight fractionation effects during their northward migration driven by LRAT. The source of HCHs was determined via the ratios of α-HCH/γ-HCH, all of which were less than 4, suggesting that the composition of HCHs was mixed with lindane (Cai et al., 2010). In Arctic sea water and sea ice meltwater, α-HCH contributed predominantly to the total OCPs, with concentrations ranging from 224 to 253 pg/L and 34.7 to 48.2 pg/L, respectively (Bigot et al., 2017).

As for DDTs, a bell shape trend was found in the Chukchi Sea and Bering Sea from1988 to 2008 alongside the ascending trend in the Sea of Japan. Geographical differences could be the reason for this temporal trend (Cai et al., 2010). Average concentrations of ΣDDTs ranged from 0.11 to 0.33 pg/L in East and West Lakes, from 0.21 to 0.49 pg/L in the Barrow Strait, while higher concentrations detected in rivers ranged between 1.40 to 2.78 pg/L, possibly relating to higher sediment loads. Moreover, HCHs and DDT in snowmelt waters were far higher in West River without obvious seasonal differences in East River (Cabrerizo et al., 2019).

Data on other OCPs rather than for HCHs and DDTs was limited. In some studies (Cai et al., 2010; Chernyak et al., 1995; Yao et al., 2002), dieldrin and endosulfan I were identified for the first time in Chukchi Sea and Bering Sea (Fig. S2, Table S9 - S10). Their mean concentrations were 52.29, 39.67, 100.67 pg/L in Bering Sea and 28.00, 32.50, 67.00 pg/L in Chukchi Sea, respectively. Dieldrin was also a significant organochlorine contaminant which has been detected in most of the matrices in the Arctic, with concentrations ranging from <LOD to tens of pg/L in water matrices. They were consistent with the results of several previous studies on OCPs in sea water from polar regions (Bigot et al., 2017). Aldrin, heptachlor and heptachlor epoxide were also observed in the polar areas. Heptachlor, with average concentration up to 94.54 pg/L was detectable in 47% of stations in Chukchi Sea. Heptachlor epoxide, a metabolite of heptachlor, was reported with average concentration up to 80.9 pg/L in Chukchi Sea (Yao et al., 2002). The wide-spread acceptable theory with regard to the global distribution of OCPs was that the main sources of OCPs emissions were in the tropics, with concentration changes in a gradient from south to north.

6.2.3. Soil and sediment

Soil has been regarded as a primary reservoir of semi-volatile organic pollutants including PCBs and OCPs, playing an important role in their distribution in the atmosphere (Cui et al., 2020). HCHs (α-HCH, γ-HCH), and DDTs were the predominant OCPs compounds measured in most of the soil samples, with concentrations ranging from MDL to 1.54 ng/g dw for HCHs, 0.34 to 19.5 ng/g dw for DDTs, respectively (Fig. S3; Table S11). Casal et al. (2018) reported that HCB was the main contributor (96 ± 3.9%) to OCPs, with soil concentrations ranging between 0.61 to 1.5 ng/g, dw. Concentrations of OCPs did not present a a clear tendency for seasonal variation. In contrast, obvious temperature-related seasonal changes were found among the ambient air and soil (or snow) fugacity of HCHs and HCB (Casal et al., 2018).

The resuspension of sediments results in the release organic pollutants back to the aquatic environment, serving as a secondary source of OCPs, which could pose an influence on their cycling in the environment (Palm et al., 2004). The most abundant OCPs were HCHs and DDTs in Arctic sediments with average concentrations ranging from 0.2 ~ 4.58 ng/g dw to 0.8 ~ 6.9 ng/g dw respectively (Fig. 4; Table S12).

Compositional patterns of HCHs and DDTs suggested that they are primarily derived from early residues of river runoff. HCB was also detected in Arctic sediments with concentrations between MDL to 3.39 ng/g dw. The highest concentration was in the Bering Sea (3.39 ng/g dw) which showed a decreasing trend over time. The concentrations of total OCPs (HCB, HCHs, chlordane, endosulfan, ∑DDTs and dieldrin) in the sediment of the Bering Sea (13.37 ng/g dw) were slightly higher than those in the Chukchi and adjacent Arctic Ocean (11.915 ng/g dw) in 2010. Deposition rates were high in the northern Bering and Chukchi Seas, while much lower in the central Arctic Ocean (Darby et al., 2009). Therefore, marine sediments in the Arctic coastal shelf regions with the highest deposition rates were likely to be important reservoirs of POPs (Kahkashan et al., 2019).

Some studies have demonstrated that POPs stored in suspended particulate matter (SPM) in glaciers and rivers, whose catchments could be discharged into the receiving marine environment can be secondary discharge sources (Carlsson et al., 2018; Garmash et al., 2013; Kallenborn et al., 2012). HCB concentrations in marine sediments (0.192 ~ 0.687 ng/g dw) were higher than those in river suspended particles (0.016 ~ 0.073 ng/g dw), as Fig. 4 shown. Furthermore, terrestrial particles with low pollutant concentrations were likely to be regarded as adsorbents for dissolved pollutants in coastal environments, thereby affecting the bioavailability of POPs to the marine food web and resulting in lower concentrations in coastal sediments and waters (Johansen et al., 2021).

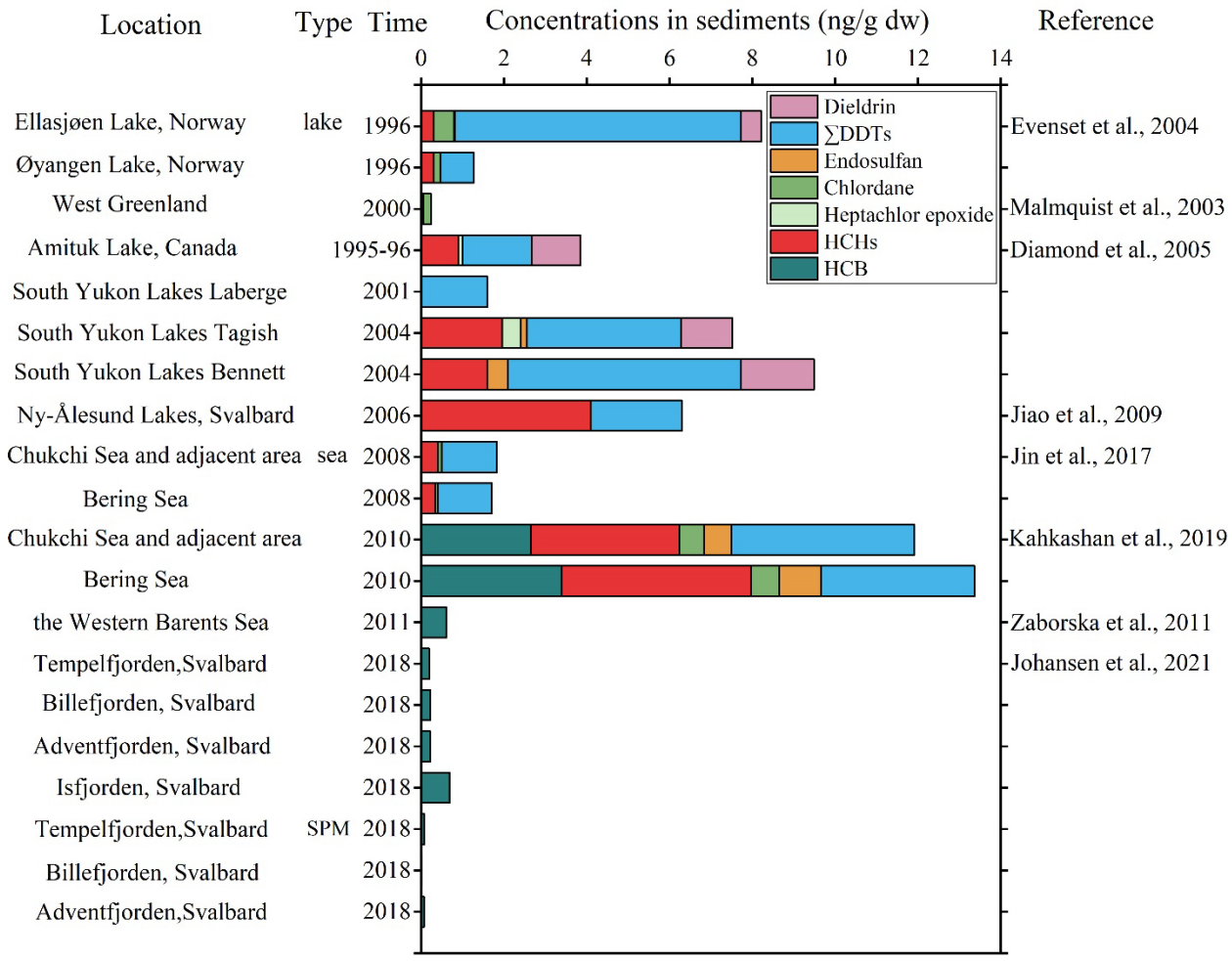


Fig. 4. Average concentrations of OCPs (ng/g dw) in sediments and SPM in Arctic areas. The original data was retrieved from a range of studies, whose references were listed in Table S12 of Supporting Information.

6.2.4. Vegetation and biota

Despite the importance of OCPs as major ecosystem contaminants, there is lack of information about their presence in soils and vegetation in circumpolar Arctic regions. Vegetation, including lichens and mosses, has been the main focus of limited studies of terrestrial food webs in the Arctic (Fig. S4; Table S13). Cabrerizo et al. (2018a) was the first study since 1999 ~ 2002 to report the residual of PCBs and OCPs in lichens and vegetation in Arctic. Consequently, gaps in current and historical data on POPs concentrations in Arctic terrestrial vegetation have been filled. HCHs, the main component of OCPs, had

454 an average concentration range of 0.06 ~ 8.59 ng/g dw before 2000 (AMAP, 2004), whereas Cabrerizo et
455 al. (2018a) measured an average concentration range of 0.07 ~ 1.39 ng/g dw in Melville Island and
456 Cornwallis Island from 2015 to 2016. The average concentration of DDTs ranged from 0.03 ~ 2.94 ng/g
457 dw prior to 2000 (AMAP, 2004), while the average concentration declined ranging from 0.007 ~ 0.22 ng/g
458 dw (Cabrerizo et al., 2018a). The average concentration of HCB (0.13 ~ 0.59 ng/g dw) was comparable
459 across recent studies. The most enriched PCB congeners and OCPs compounds in vegetation were the low
460 molecular weight PCBs and OCPs, proving the significance of long-range atmospheric transport (Cabrerizo
461 et al., 2018a).

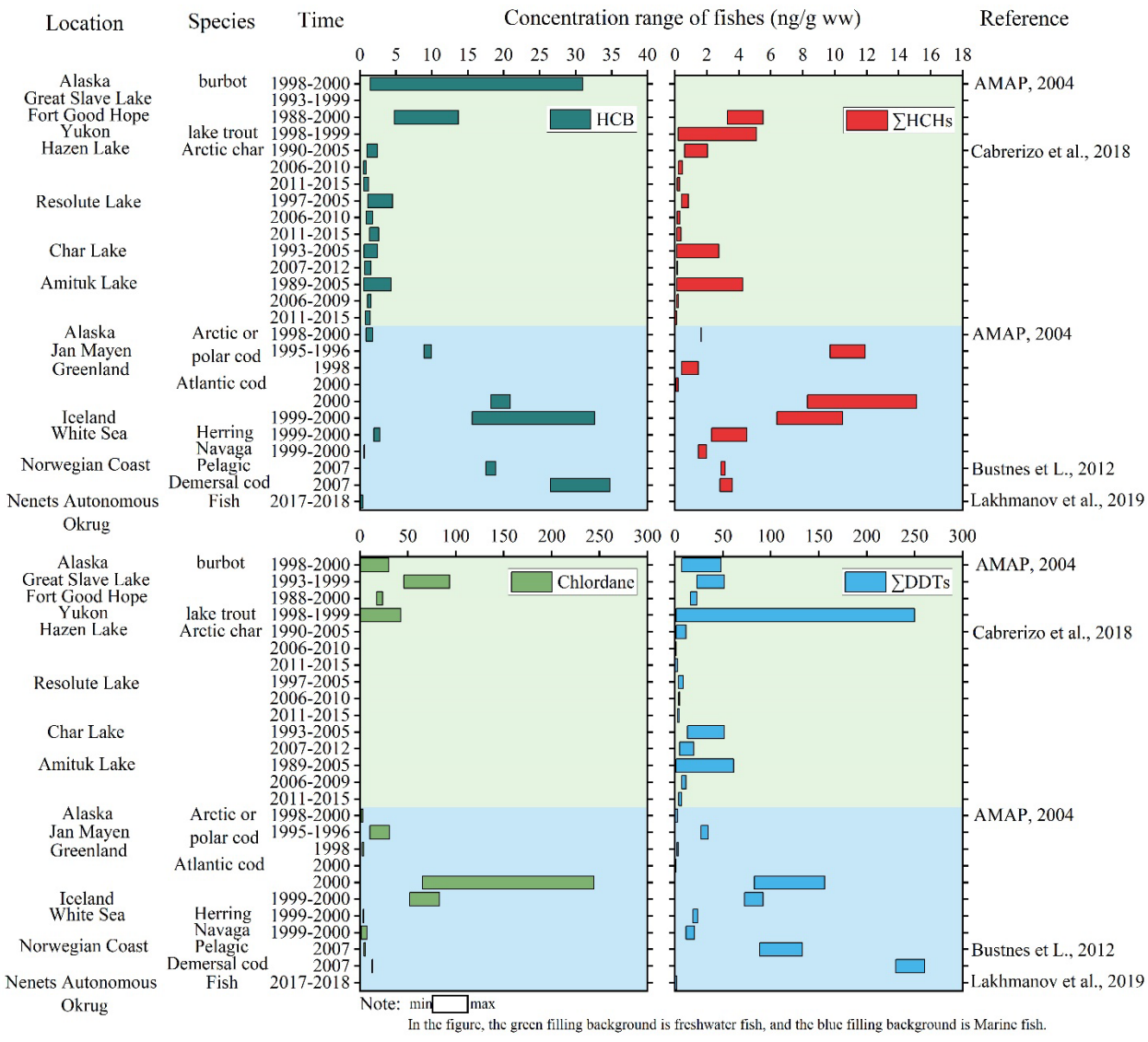
462 Animals are also important parts of the ecosystem. Keeping track of OCP concentrations across diverse
463 species of animals is important to explore the trends of legacy of pollutants, which is of great significance
464 to the improvement of ecological environment and human health. The main POPs reported in fish were
465 DDT and its degradation products, HCB and HCHs were also frequently detected in fish in both fresh and
466 sea water. Among the target compounds for OCPs in Arctic char caught from four lakes (Amituk, Resolute,
467 Char and Hazen Lakes), toxaphene always displayed the highest concentrations, followed by ΣDDTs and
468 ΣHCHs, whose average concentrations varied from 1.6 to 172 ng/g ww (wet weight) for toxaphene, from
469 0.54 to 61 ng/g ww for ΣDDT and from 0.03 to 4.24 ng/g ww for ΣHCHs, respectively (Fig. 5; Table S14).
470 ΣDDTs were found to have declined in four lakes (Amituk, Resolute, Char and Hazen Lakes), with
471 proportion from 7.1%/yr to 11%/yr. Meanwhile, α-HCH decreased remarkably in all lakes as well, with the
472 rate of 7.8%/yr ~ 14%/yr. HCB also dropped at a rate of 4%/yr. However, β-HCH did not exhibit any
473 considerable decrease. Toxaphene concentrations only reduced in Amituk Lake and Lake Hazen at the rate
474 of 6.4%/yr and 7%/yr, respectively (Cabrerizo et al., 2018b). In sea water, the concentrations of OCPs were
475 two to three times higher for cod than for pelagic saithe on the Norwegian Coast, especially for highly
476 halogenated-substituted compounds. It is likely that they bioaccumulate through benthic organisms rather
477 than pelagic food chains, possibly on account of the adsorption of higher molecular weitght compounds on
478 bottom sinking particles (Bustnes et al., 2012). Moreover, it has been shown that the composition of

contaminants in cod changed gradually from south to north, particularly for HCB, chlordane and ΣDDTs in the contaminant burdens of cod applied with latitude, whereas the latitudinal fractionation was insignificant in saithe, which was likely resulting from its pelagic and nomadic behavior (Bustnes et al., 2012). In addition, OCPs, such as DDTs, were highly positively correlated with lipid content (Lakhmanov et al., 2020).

Concentrations of OCPs in Arctic birds were in the range of < LOD to tens of thousands of ng/g (Fig. 6; Table S15). The OCPs levels in the species of Arctic seabirds were the highest and it was identified as a vital stressor in a species already at risk owing. For example, the concentrations of OCPs in ivory gull eggs were more than 30 000 ng/g lipid weight (lw) in the Russian Arctic in 2007 (Miljeteig et al., 2009). HCB, chlordane, DDTs, dieldrin and mirex in eggs of thick-billed murres, northern fulmars and black-legged kittiwakes exhibited significant decreases or no prominent variation between 1975 and 2003 measured from Prince Leopold Island of the Canadian High Arctic. Nevertheless, significant increases were found for ∑HCH in the kittiwakes and fulmars (Braune, 2007). The main component of DDTs was DDE, and its presence in birds showed an annually decreasing trend. High levels of OCPs were observed in all tissues, except in adipose tissue. Eggs generally exhibited the highest concentrations, with muscle containing relatively high levels while adipose tissue displayed the lowest levels both on a lipid weight and a wet weight basis. More than a hundredfold higher variation in contamination loads was reported amongst individuals. The variations in concentrations between males and females were not remarkable, but the discrepancies between juveniles and adults were significant (Jaspers et al., 2013). These findings suggested that birds can be useful bio-indicators providing high spatial resolution for contaminant source apportionment in Arctic terrestrial environments.

Reindeers, serving as a valuable bioindicator, were the only long-lived vegetarian mammals feeding on local vegetation, being exposed to contaminants mostly via diet of different kinds of locally grown lichen and moss. In regard to 15 interested pesticides, only DDT and its metabolites exceeded the detectable limit, with the highest detectable value of *p*,*p*'-DDD (57.5 ng/g dw) in one sample from Longyearbyen. However,

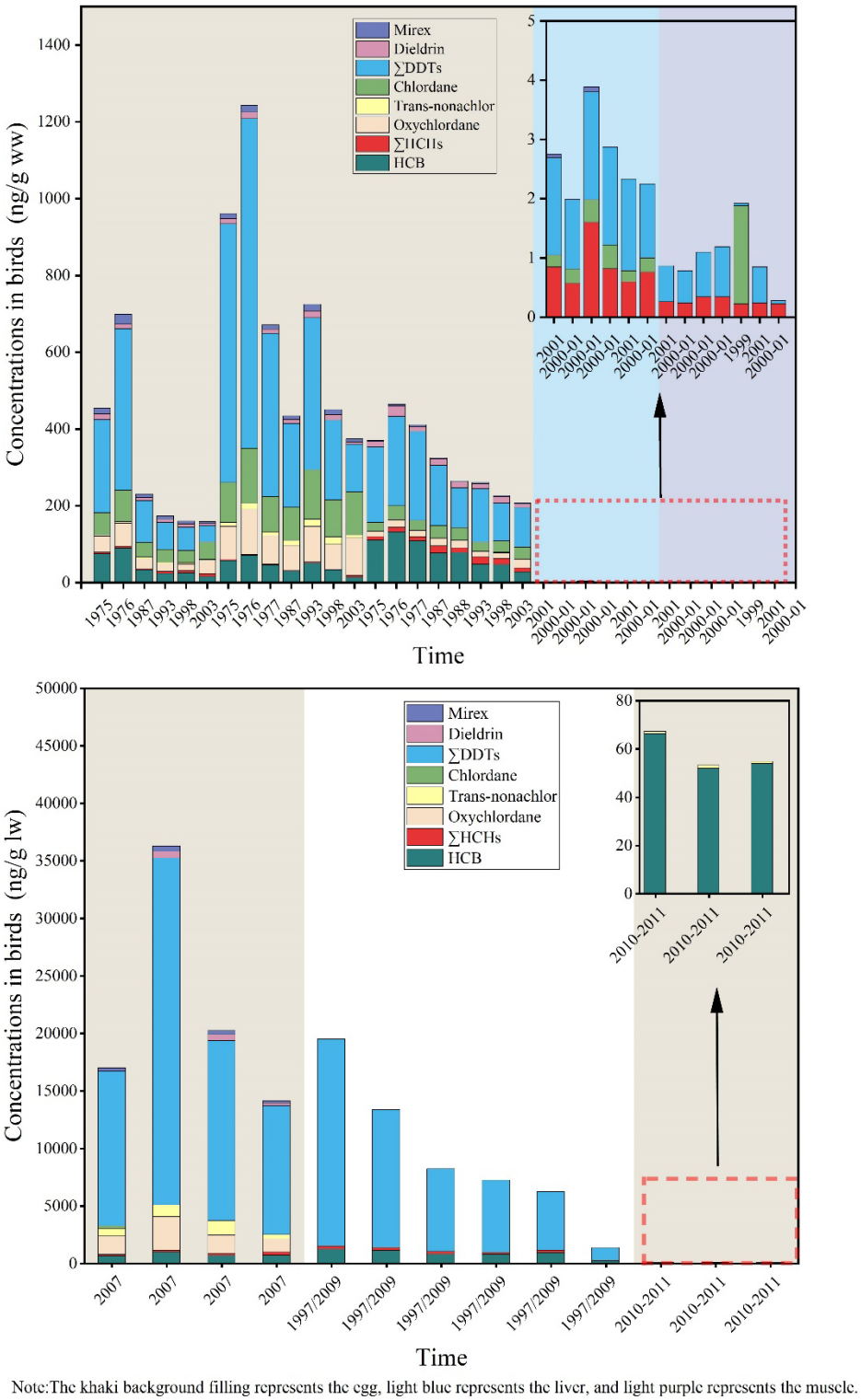
504 despite this single outlier, *p*,*p*'-DDD concentrations in the remaining samples were much lower (median
505 1.44 ng/g dw) (Pacyna-Kuchta et al., 2020). Some OCPs were detectable in human plasma in the
506 Arctic/subarctic region of mainland Russia, mainly including PCBs, *p*,*p*'-DDE, *p*,*p'*-DDT, *β*-HCH, and
507 HCB, with those in the Aral Sea region of Uzbekistan serving as a control group. The contamination in
508 human plasma can be explained by local consumption of marine mammals, as well as the recent use of
509 pesticides. Polar bears in the western Russian Arctic had the highest concentrations of chlordane and *p*,*p*'-
510 DDE, compared with polar bears in the western and eastern regions. There were regional differences in
511 OCPs concentrations, which indicated differences in contamination sources and dietary habits. Furthermore,
512 it inferred the existence of an important source in the Russian Arctic (Lie et al., 2003). As rising
513 temperatures in the Arctic have created new communities that attract new predators such as whales, the
514 levels of POPs in Arctic whales should be monitored continuously to provide long-term data. Tartu et al.
515 (2020) reported that DDTs and PCBs were the major POPs in fin whales and blue whales in Svalbard,
516 Norway, and the concentration of pollutants in fin whale was 1.6 ~ 3 times higher than that in blue whale,
517 which may be due to the higher trophic level of fin whale. There were fewer available concentration data
518 on OCPs in Arctic invertebrates (Table S17). Studies have shown that macroinvertebrates such as
519 *Gammarus* species and molluscs have been used in the past for biomonitoring of POPs, but the former have
520 been mainly associated with metals, whereas the latter's OCPs concentrations in the body do not correlate
521 with some of the OCPs concentrations in the sediment. This is probably as result of their lower position in
522 the food chain (Lebrun et al., 2015; Lu et al., 2017). McGovern et al. (2022) observed indications of
523 accumulation of remobilized *α*-HCH in coastal zooplankton in the summer, but the resulting concentrations
524 were low, which also suggested that glacial meltwater was a secondary source of *α*-HCH to zooplankton.
525 Invertebrates play an important role in the stability of freshwater and marine ecosystems. Glacial melting
526 and terrestrial inputs can affect the distribution of contaminants in the environment, and future research
527 should be carried out on the effects of a combination of factors on the fate of OCPs in invertebrates.



528

529 **Fig. 5.** The concentrations of OCPs in fishes (ng/g) in the Arctic. The original data was retrieved from a

530 wide range of studies, whose references were listed in Table S14 of Supporting Information.



531

532 **Fig. 6.** The concentrations of OCPs in birds (ng/g) in the Arctic. The original data was retrieved from a

533 wide range of studies, whose references were listed in Table S15 of Supporting Information.

7. Current-use pesticides

Since the legacy OCPs have been banned or restricted for use in recent years, current-use pesticides (CUPs) have served as substitutes as CUPs are thought to be less persistent in environment and safer for biota. CUPs contain more than 900 active ingredients, involving organophosphate pesticides, pyrethroids, carbamates, neonicotinoids, and sulfonylureas (Wang et al., 2018). Recently, however, several studies have demonstrated that CUPs undergo long range transport from source regions to pristine polar areas through air mass transport (Matthies et al., 2009; Pućko et al., 2017; Scheringer et al., 2009; Stocker et al., 2007).

The research on CUPs in polar areas are rather limited, albeit attracting worldwide attention. Thus, the transport paths of CUPs as well as the role of polar areas in their global distribution still remains uncertain. Balmer et al. (2019) have reviewed the presence of CUPs in Arctic areas. Three compounds, dacthal, chlorothalonil, and endosulfan, were the most widely reported CUPs in Arctic air. Passive sampling methods were used to monitoring CUPs, with the absorption phase of PUF, XAD or PUF-XAD sandwich. The concentrations of α-endosulfan (at the level of several or 10's of pg/m^3) were the highest among the detectable CUPs (Baek et al., 2011; Bossi et al., 2013; Bossi et al., 2016; Jantunen et al., 2015; Pozo et al., 2006; Wania et al., 2003). CUPs in air samples collected on a scientific cruise stretching from the East China Sea to the High Arctic demonstrated that the Artic could be influenced by direct input from Russia and North America (Balmer et al., 2019; Zhong et al., 2012).

In surface seawater from the Sea of Japan into the Chukchi Sea and western Arctic Ocean, α-endosulfan, β-endosulfan, endosulfan sulfate and methoxychlor were the most detected CUPs with the concentrations range of < MDL to 10's of pg/L (Zhong et al., 2012). However, concentrations of endosulfan sulfate and dacthal were the highest and the most frequently measured CUPs in the dissolved phase, followed by chlorpyrifos, pentachloronitrobenzene (PCNB) and α-endosulfan in the Canadian Arctic. (Morris et al., 2016). Pućko et al. (2017) discovered that melt ponds in Canadian Arctic Archipelago might play an important role in the delivery of CUPs to the surface layers of the Arctic Ocean, based on the result that the concentrations of 4 CUPs (chlorpyrifos, dacthal, PCNB and trifluralin) in snow-melt ponds were

559 10 ~ 16 folders higher of those in sea water. On the contrary, chlorothalonil and α-endosulfan were not
560 found in the ponds, suggesting they either volatilized into the atmosphere or were flushed into the ocean
561 before the snowmelt began (Balmer et al., 2019). Moreover, the runoff of glacial melt water had an impact
562 on the levels of α-endosulfan in surface sediments of a glacial fjord in Svalbard, Norway (Balmer et al.,
563 2019; Ma et al., 2015).

564 By reviewing studies on the temporal variation of CUPs in Arctic areas, it was demonstrated that the
565 concentrations of CUPs in sea water and air have slightly declined or remained stable over recent times.
566 Specifically, atmospheric concentrations of α-endosulfan at Alert have declined slightly, while in Fox lake
567 (Canada) they remained relatively constant (AMAP, 2014; Yu et al., 2015). Nevertheless, in surface sea
568 water collected in the Canadian Arctic Archipelago from 2011 to 2013, concentrations of several CUPs
569 increased, although there was no obvious temporal change in the concentrations of other CUPs (chlorpyrifos,
570 dacthal) (Balmer et al., 2019; Jantunen et al., 2015).

571 Due to the lack of the research on CUPs in vegetation, fish and mammals in the Arctic, CUPs
572 biomagnification and transport through food web were remains difficult to determine. CUPs were detected
573 in a variety of plants in Arctic Canada, such as lichens, moss and cotton grass. The concentrations of CUPs
574 in vegetation were higher than those in caribou (*Rangifer tarandus*), and wolves (*Canis lupus*), suggesting
575 that no bio-magnification existed (Morris et al., 2016). In order to discern the trophic dilution or trophic
576 magnification of CUPs, more biota data are required.

577 8. Ecological risk assessment and biological amplification

578 *8.1. Ecological risk assessment*

579 Sediments and soils are essential pathways for aquatic and terrestrial biota to be exposed to PCBs and
580 OCPs. Two indicators, interim sediment quality guidelines (ISQGs) and probable effect levels (PELs)
581 which were set in accordance with the specifications described in Canadian Council of Ministers of the
582 Environment (CCME) (Gaudet et al., 1995) can be used for ecological risk assessment of sediments. A
583 value less than ISQG indicates no risk, a value greater than PEL indicates a possible risk, and a value

584 between ISQG and PEL represents an occasional impact (Kahkashan et al., 2019). The mean concentrations
585 of PCBs in sediments at all sites in Table S5 did not exceed the PCBs ISQG value (34.1 ng/g in freshwater
586 sediments and 21.5 ng/g in marine sediments), except for Lake Ellasjøen (Bjørnøya, Norway) and Ebbaelva
587 River (Isfjorden, Svalbard), where the average PCBs concentrations were 46.4 ng/g and 582.89 ng/g,
588 respectively. The concentration of PCBs in Lake Ellasjøen did not exceed the PEL value of PCBs (277
589 ng/g), but the highest concentration of PCBs in Ebbaelva River (Isfjorden, Svalbard) reached 2800 ng/g,
590 indicating that there may be a possible ecological risk in this area and further studies on sources of
591 contaminants and sediment quality improvement programs in the area are required. According to the studies
592 we reviewed (Table S12), chlordane concentrations were all below the ISQG value (2.26 ng/g), indicating
593 no biological effect risks. The concentrations of most DDTs did not exceed the ISQG value (4.48 ng/g), but
594 some exceeded the ISQG value but fell below the PEL value (386.28 ng/g) at sites Ellasjøen Lake (6.9 ng/g,
595 Norway) and South Yukon Lakes Bennett (5.63 ng/g). The comparison between concentrations of HCHs
596 and the PEL indicate that there may be an occasional impact in the study area.

597 Canadian Soil Quality Guidelines (SQG_S) were developed by integrating both human health and
598 ecological environmental impacts. The human health soil quality guidelines (SQG_{HH}) and the environment
599 soil quality guidelines (SQG_E) were formulated respectively, and the lower of the two was taken as the soil
600 quality guideline value (SQG_S). Within this guideline, the agricultural land standard was the highest, so the
601 agricultural land standard was used for ecological risk evaluation. The mean concentration for PCBs in soil
602 in Table S4 did not exceed the SQG_S value (500 ng/g), but higher concentrations were reported at individual
603 sampling sites, such as Barentsburg (Russia) (28700 ng/g) and Pyramiden (Russia) (13900 ng/g). The
604 potential reason was that the sampling sites were located in a coal mining settlement, where the paint (3520
605 mg/kg) and small capacitors (114,000 mg/kg) had very high concentrations of PCBs, suggesting that these
606 two were the main sources of local PCBs pollution (Jartun et al., 2009). None of the average DDT soil
607 concentrations shown in Table S11 were higher than the SQG_S value (700 ng/g). The ecological risk
608 assessment of PCBs and OCPs in soil was also compared with the Chinese Soil Environmental Quality

609 Standard (GB15618-2008) (Zheng et al., 2022) which is more stringent than the Canadian standard, and
610 again no ecological risk of PCBs and OCPs was apparent.

611 Even though the average concentration of PCBs in soil shown in Table S4 did not exceed the level I
612 threshold value (15 ng/g), the highest concentrations at some sampling sites did, although they did not
613 exceed the secondary criterion (100 ng/g) for agricultural land (except for Barentsburg, Pyramiden, and
614 Cape Dyer (591 ng/g, Canada). Both HCHs and DDTs in soil did not exceed the first level standards (HCHs
615 is 10 ng/g, DDTs is 50 ng/g), except for DDTs in Alaskan soil from the United States which reached 19.5
616 ng/g dw during 1991 ~ 1993 (Table S11). Combining the two methods for assessing the impacts of PCBs
617 and OCPs on human health and the ecological environment, we found that these two groups presented no
618 risk on human health and the ecological environment across the Arctic, with the exception of a few sampling
619 areas that were potentially associated with nearby industrial production.

620 Plasma concentrations of PCBs in people from Arctic regions have been investigated for several
621 decades as PCB exposure occurs via the food web. Owing to dietary habits similar to those of marine
622 mammals, plasma levels of PCBs have been consistently high in circumpolar residents, especially in East
623 Greenland, where PCBs concentrations in blood have exceeded Canadian standards (Deutch et al., 2007).
624 Total PCB concentrations in the blood of pregnant females in Nunavik declined by 84% from 1992 to 2017.
625 However, 10% of the plasma samples of women were above the guideline values in 2017. Despite the
626 marked reductions in PCBs levels, the exposure to these contaminants remained quite common among
627 pregnant women in Nunavik (Adamou et al., 2020). Many studies have confirmed that OCPs can cause
628 endocrine disorders and immune dysfunction in birds. Some OCPs delayed the time of laying and increased
629 the number of non-viable eggs (Miljeteig et al., 2012). In future studies, we should strengthen the
630 monitoring of new pollutants, such as CUPs, antibiotics and microplastics, establishing concentration
631 thresholds that may pose risks to better assess their impact on humans and the entire ecosystem.

8.2. Biotransfer pattern through food web

In the Arctic ecological environment, PCBs and OCPs are transferred through the food web, continuing to accumulate and concentrate. This effect can lead to high concentrations of pollutants in top predators at higher TLs, ultimately endangering biodiversity as well as human health (Binnington et al., 2016; Nuijten et al., 2016; Szczybelski et al., 2016). Pedro et al. (2017). Atkinson et al. (2019) found very high concentrations of PCBs and OCPs (ΣCHLs and ΣDDTs) in biota across Greenland, Denmark and Russia. The average lipid concentration was 40 to 70 μg/g, with the exposure threshold of organic halogenated pollutants in Arctic organisms estimated to be at the 1 μg/g level, with the concentration in killer whales exceeding the threshold by dozens of times. A series of studies have shown that biomagnification is more evident in polar areas. Verhaert et al. (2017) elucidated that the TMFs for PCBs in Arctic food webs were higher than those in subtropical aquatic food webs. Kelly et al. (2009) discovered that in the Arctic marine mammal food web, the concentrations of Perfluorooctane Sulfonamide (PFOSA) and lipophilic organochlorine compounds increased with the increase of TLs, with PCB153 and *p,p*'-DDE showing obvious biomagnification. Hobson et al. (2002) studied the *in vivo* transformation behavior of PCBs, establishing the linear relationship between pollutant concentration and $\delta^{15}N$, demonstrating the biomagnification process of PCB153 in Arctic food webs. Corsolini and Sarà (2017) built the relationship between TLs and concentrations, which demonstrated the biomagnification of polar PCB and DDE by calculating the TMFs.

Data were collected on concentrations of PCBs and OCPs in Arctic fish, birds, and mammals, as well as TLs for each species. According to the previous studies (Alava and Gobas, 2016; Fisk et al., 2001; Hallanger et al., 2011; Kelly et al., 2009; Verhaert et al., 2017), the relationship between the $\log_{10}$-transformed compounds concentrations and TLs has been established. In addition, TMFs were calculated by $\log_{10} C = a \times TL + b$, $TMFs = 10^a$. As shown in Figs. 7 and Fig. S5, the concentrations of HCB and ∑HCHs increased significantly with TLs, indicating a significant positive linear correlation ($R^2 = 0.6203$, $R^2 = 0.6836$; $P < 0.001$), while ∑DDTs increased moderately with TLs, exhibiting a significant relationship

($R^2 = 0.4314$; $P < 0.001$). Chlordane, mirex, and ∑PCBs concentrations also increased with TLs. Although their R^2 was greater than 0.2 and less than 0.3, their sample sizes were large with significant correlations between $\log_{10}$-transformed concentrations and TLs ($P < 0.001$). The differences in the relationships between pollutant concentrations and TLs might be caused by the discrepancy of the contaminants physicochemical properties, the distinction in feeding habits of diverse species of organisms, and the diversity in pollutant absorption and enrichment capacity (Masset et al., 2019). However, there was no significant relationship between the concentration of dieldrin and TLs ($R^2 = 0.1156$; $P = 0.089$), which might be due to the limited concentration data associated with dieldrin and the narrow range of biotrophic levels involved. Calculation of the TMFs are depicted in Fig. 8, with TMFs all over 1, except for dieldrin, ranging from 6.33 to 26.64. These data indicate that biological magnification of HCB, ∑HCHs, ∑DDTs, chlordane, mirex, and ∑PCBs occurred to certain extent. This observation was consistent with the results of previous studies. For example, Kelly et al. (2008) reported that the TMFs for PCBs in invertebrates, fish, seals, and polar bears varied from 2.9 to 11. Muir et al. (2003) provided the TMFs of OCPs for invertebrates, fish and seals which ranged between 0.66 to 9.6 (chlordane) and for PCBs between 0.8 to 3.4. The differences in TMFs are due to varying concentrations of OCPs and PCBs in Arctic fishes, birds, and mammals collected in this review, which covered a wider range with higher TLs, making it more meaningful to analyze biomagnification through food webs. This may also be the reason for the relatively high TMF values in this study. Biomagnification is associated with the interactions of a range of factors. For example, the greenhouse effect influenced the melting of Arctic glaciers to accelerate the release of POPs into the food web (Amiraux et al., 2023). Moreover, biological invasion (Frossard et al., 2023), ecological characteristics of organisms (respiratory elimination), feeding habits (Masset et al., 2019), the long half-life of POPs, their biomagnification potential, and so on, all had impact on the process (Evenset et al., 2016). It has also been discovered that the K_{OW} and K_{OA} of compounds also altered biomagnification. The pollutants with lower K_{OW} and higher K_{OA} were more likely to bioaccumulate (Moses et al., 2015). Benthos was found to contribute a lot to higher-trophic level organisms (Amiraux et al., 2023), and more effort should be made

to understand the impact of phytoplankton and animals on higher trophic species. Hallanger et al. (2011) confirmed that zooplankton are able to magnify POPs, which generated a biological amplification effect as a crucial food source for fish. Recent studies have found that bioaccumulation of Microplastics (MPs) was found in many zooplankton functional groups in the oceans (Aytan et al., 2022; Botterell et al., 2019; Goswami et al., 2023). Xu et al. (2020) collected 38 species of invertebrates, and suspect microplastics were found in 32 species of organisms. MPs could also be vectors of hydrophobic organic pollutants, such as PCBs and OCPs owing to their large surface area to volume ratio and lipophilicity. The effects of MPs on the enrichment and biotransformation of PCBs and OCPs in Arctic organisms have not been reported, and future research on the impact of MPs on PCBs and OCPs in polar regions should consider these factors.

The biomagnification of OCPs and PCBs ultimately affects biodiversity and has a profound effect on the health of top predators, including humans. Meanwhile, residents, especially those living in polar regions, can confront lower health risks by adjusting dietary strategies, such as reducing the consumption of seafood from highly contaminated areas. In the future, it is expected to conduct more in-depth research on the Arctic ecological environment, put forward more valuable propositions for the conservation of the polar regions even the global ecological environment.

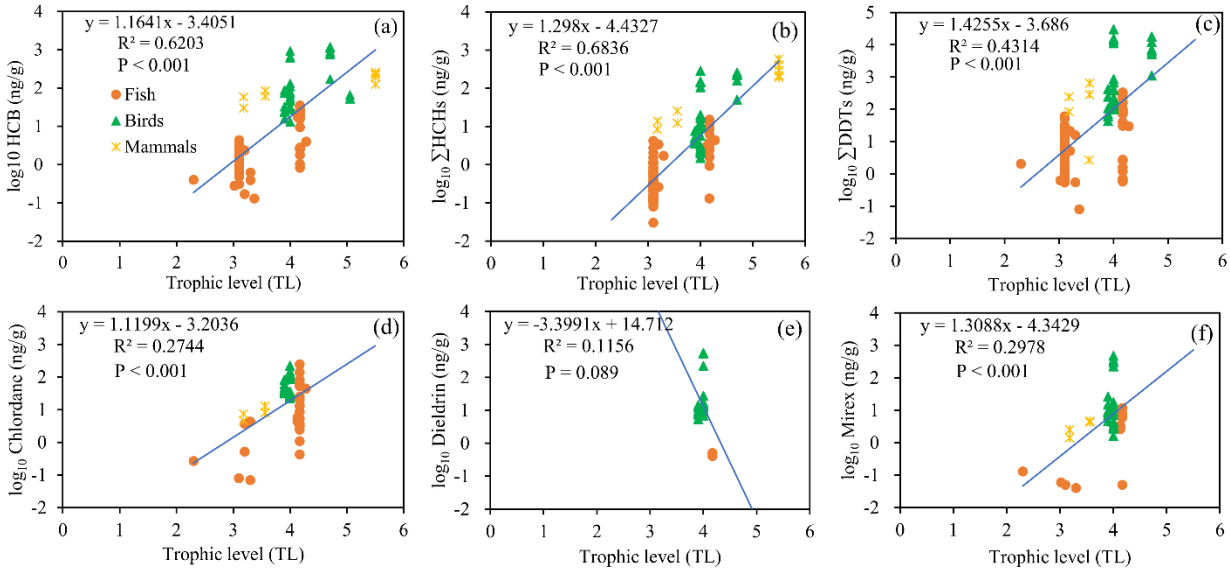


Fig. 7. The relationship between log_{10}-transformed concentration for selected OCPs (ng/g) and trophic level (TL) in fishes, birds, and mammals in the Arctic.

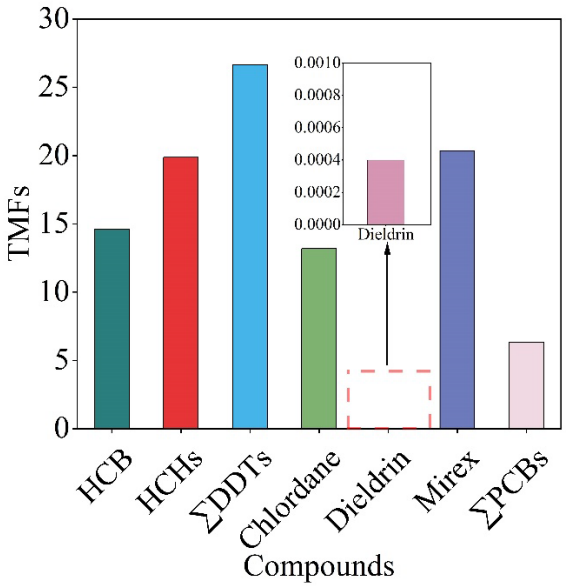


Fig. 8. Trophic Magnification Factors (TMFs) for OCPs and PCBs in Arctic trophic webs.

9. Conclusions and perspectives

The presence of POPs in the Arctic has been of global concern in recent decades and a great deal of effort has been devoted in exploring their environmental behavior. By reviewing a hundred current pieces of research, the main purpose of this study was to discuss their legacy, sources, environmental behavior and risks. On a global scale, PCBs and OCPs from Eurasia and Northern America were transferred into the Arctic areas through air mass movement. However, river runoff inputs, animal migration, glacier melt also played key roles in specific locations, such as Bering seas of Russia, Norway, and Canada. On the basis of more than two decades of scientific investigation on PCBs and OCPs in the Artic atmosphere, it is clear that PCBs and OCPs concentrations have been gradually declining. After 2015, the concentrations of HCB, HCHs and chlordane in the atmosphere have reduced by one to several orders of magnitude compared with those before 2000. In aqueous samples, concentrations of PCBs and OCPs increased dramatically during non-glacial periods, suggesting that snowmelt water in these countries was the source to nearby rivers. PCBs were more abundant in Arctic vegetation than in soil and sediment, with concentrations several higher

than in the latter. The most abundant OCPs in soil were HCHs and DDTs, although there was no seasonal trend in concentrations. The contribution of HCHs and DDTs to OCPs was higher in sediments, which were mainly derived from river runoff. PCBs and OCPs in the Arctic areas undergo dynamic cycling between air, soil and water with sinks and sources swapping places. Levels of PCBs and OCPs in biota were higher than those in environmental media, especially in seabirds and mammals with high TLs. Although PCBs and OCPs are banned, some emerging unintentionally produced PCBs and CUPs are still detected in the Arctic, which require continuing assessment of their human health and ecological risk.

The ecological risk assessment of soil and sediment was carried out by combining the Canadian Soil Quality Guidelines and Chinese Soil Environmental Quality Standard (GB15618-2008). With the exception of occasional high concentrations, neither PCBs nor OCPs in soil and sediment exceeded the values of these two standards, indicating that there was no ecological risk in Arctic soil and sediment. In order to analyze the biological effects of PCBs and OCPs, the relationship between TLs and $\log_{10}$-transformed compound concentrations in fish, birds and mammals was established. The concentrations of HCB, ∑HCHs, ∑DDTs, chlordane, mirex, and ∑PCBs increased with TLs, suggesting a significant positive linear correlation. This confirms the biological magnification of HCB, ∑HCHs, ∑DDTs, chlordane, mirex, and ∑PCBs. The biomagnification of OCPs and PCBs ultimately disrupts biodiversity and has a profound effect on the health of top predators, including humans. Consequently, we need to continue to pay close attention to the presence of PCBs and OCPs residues in polar areas.

(1) Laboratory detection methods need to be further developed to cover a wider range of target compounds. Some UP-PCBs, account for a relatively higher proportions of totals, and some degradation products have not been fully understood yet, and maybe more toxic. The optimization of analysis methods and more advanced instrumentation are the prerequisite for accurately determining their concentrations. Therefore, the analytical capacities need be strengthened in the future.

(2) Tissue-specific protein and lipid content as well as many other biochemical processes are of significance to the toxicokinetics and fate of metabolites and natural products of chlorinated contaminants.

Therefore, the selective retention of certain organohalogens in specific tissue/body parts are likely to make contributions to site-specific toxicological effects on heavily contaminated species such as Arctic white gull. Detailed studies are required to elucidate processes and mechanisms of macromolecular binding interactions and accumulation of organohalogens such as OCPs and PCBs in birds and other Arctic species. In addition, MPs can act as carriers of PCBs and OCPs, and the effects of MPs on the accumulation and biotransformation of PCBs and OCPs in Arctic organisms has not yet been reported, and future research on the combined effects of some new environmental factors, such as MPs, on PCBs and OCPs in polar regions is required.

(3) It is important to set up concentration thresholds for individual compounds and mixtures for the protection of polar ecosystems. For one thing, there is a necessity to identify and refine concentration thresholds for biologically related health effects for polar wildlife and fish. For another thing, there is a necessity to better characterize the cumulative impacts and interaction thresholds for contaminant exposure. This facilitates a more accurate and comprehensive assessment of the ecological risks arising from contaminants.

(4) It is important to clarify the relationship among the food web with high trophic position species and pollutants, to use and optimize multiple models to quantify the relationship between the two more accurately. Factors that affect biomagnification need to be investigated in more detail and with the ecological dynamics of organic contaminants . In addition, the effects of pollutant impacts on individual organisms, species groups and communities need to be assessed to provide an understanding of the overall health of birds and mammal populations to protect biodiversity and human health.

Declaration of competing interest

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

Acknowledgment

This study was jointly supported by the National Natural Science Foundation of China (22076112 and 21976045), and China Scholarship Council (CSC) Scholarship (202308610123).

Appendix

Supporting Information to this article can be found online at……

References

Adamou, T.Y., Riva, M., Muckle, G., Laouan Sidi, E.A., Lemire, M., Ayotte, P., 2020. Blood mercury and plasma polychlorinated biphenyls concentrations in pregnant Inuit women from Nunavik: Temporal trends, 1992-2017. Sci. Total Environ. 743, 140495. https://doi.org/10.1016/j.scitotenv.2020.140495.

Addison, R.F., Ikonomou, M.G., Fernandez, M.P., Smith, T.G., 2005. PCDD/F and PCB concentrations in Arctic ringed seals (*Phoca hispida*) have not changed between 1981 and 2000. Sci. Total Environ. 351-352, 301-311. https://doi.org/10.1016/j.scitotenv.2005.04.051.

Ademollo, N., Spataro, F., Rauseo, J., Pescatore, T., Fattorini, N., Valsecchi, S., et al., 2021. Occurrence, distribution and pollution pattern of legacy and emerging organic pollutants in surface water of the Kongsfjorden (Svalbard, Norway): Environmental contamination, seasonal trend and climate change. Mar. Pollut. Bull. 163, 111900. https://doi.org/10.1016/j.marpolbul.2020.111900.

Airaksinen, R., Hallikainen, A., Rantakokko, P., Ruokojärvi, P., Vuorinen, P.J., Parmanne, R., et al., 2014. Time trends and congener profiles of PCDD/Fs, PCBs, and PBDEs in Baltic herring off the coast of Finland during 1978-2009. Chemosphere. 114, 165-171. http://dx.doi.org/10.1016/j.chemosphere.2014.03.097.

Alava, J.J., Gobas, F.A.P.C., 2016. Modeling ^{137}Cs bioaccumulation in the salmon-resident killer whale food web of the Northeastern Pacific following the Fukushima Nuclear Accident. Sci. Total Environ. 544, 56-67. https://doi.org/10.1016/j.scitotenv.2015.11.097.

Alexeeva, L.B., Strachan, W.M.J., Shlychkova, V.V., Nazarova, A.A., Nikanorov, A.M., Korotova, L.G., et al., 2001. Organochlorine Pesticide and trace metal monitoring of Russian rivers flowing to the Arctic Ocean: 1990-1996. Mar. Pollut. Bull. 43, 71-85. https://doi.org/10.1016/S0025-326X(00)00166-1.

AMAP, 2004. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic.Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2014. Trends in Stockholm Convention Persistent Organic Pollutants (POPs) in Arctic air, human media and biota.Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

Amiraux, R., Mundy, C.J., Pierrejean, M., Niemi, A., Hedges, K.J., Brown, T.A., et al., 2023. Tracing carbon flow and trophic structure of a coastal Arctic marine food web using highly branched isoprenoids and carbon, nitrogen and sulfur stable isotopes. Ecol. Indic. 147, 109938. https://doi.org/10.1016/j.ecolind.2023.109938.

Andersen, M.S., Fuglei, E., König, M., Lipasti, I., Pedersen, Å.Ø., Polder, A., et al., 2015. Levels and temporal trends of persistent organic pollutants (POPs) in arctic foxes (*Vulpes lagopus*) from Svalbard in relation to dietary habits and food availability. Sci. Total Environ. 511, 112-122. https://doi.org/10.1016/j.scitotenv.2014.12.039.

Aslam, S.N., Huber, C., Asimakopoulos, A.G., Steinnes, E., Mikkelsen, Ø., 2019. Trace elements and polychlorinated biphenyls (PCBs) in terrestrial compartments of Svalbard, Norwegian Arctic. Sci. Total Environ. 685, 1127-1138. https://doi.org/10.1016/j.scitotenv.2019.06.060.

Atkinson, S., Branson, M., Burdin, A., Boyd, D., Ylitalo, G.M., 2019. Persistent organic pollutants in killer whales (*Orcinus orca*) of the Russian Far East. Mar. Pollut. Bull. 149, 110593. https://doi.org/10.1016/j.marpolbul.2019.110593.

Aytan, U., Esensoy, F.B., Senturk, Y., 2022. Microplastic ingestion and egestion by copepods in the Black Sea. Sci. Total Environ. 806, 150921. https://doi.org/10.1016/j.scitotenv.2021.150921.

Baek, S.Y., Choi, S.D., Chang, Y.S., 2011. Three-year atmospheric monitoring of Organochlorine Pesticides and Polychlorinated Biphenyls in polar regions and the south Pacific. Environ. Sci. Technol. 45, 4475-4482. https://doi.org/10.1021/es1042996.

Balmer, J.E., Morris, A.D., Hung, H., Jantunen, L., Vorkamp, K., Riget, F., et al., 2019. Levels and trends of current-use pesticides(CUPs)in the arctic:An updated review, 2010-2018. Emerging Contam. 5, 70-88. https://doi.org/10.1016/j.emcon.2019.02.002.

Bargagli, R., 2008. Environmental contamination in Antarctic ecosystems. Sci. Total Environ. 400, 212-226. http://dx.doi.org/10.1016/j.scitotenv.2008.06.062.

Bartlett, P.W., Isaksson, E., Hermanson, M.H., 2019. 'New' unintentionally produced PCBs in the Arctic. Emerging Contam. 5, 9-14. https://doi.org/10.1016/j.emcon.2018.12.004.

Bidleman, T.F., Jantunen, L.M., Kurt-Karakus, P.B., Wong, F., 2012. Chiral persistent organic pollutants as tracers of atmospheric sources and fate: review and prospects for investigating climate change influences. Atmos. Pollut. Res. 3, 371-382. https://doi.org/10.5094/APR.2012.043.

Bidleman, T.F., Laudon, H., Nygren, O., Svanberg, S., Tysklind, M., 2017. Chlorinated pesticides and natural brominated anisoles in air at three northern Baltic stations. Environ. Pollut. 225, 381-389. https://doi.org/10.1016/j.envpol.2017.02.064.

Bigot, M., Hawker, D.W., Cropp, R., Muir, D.C.G., Jensen, B., Bossi, R., et al., 2017. Spring melt and the redistribution of Organochlorine Pesticides in the sea-ice environment: A comparative study between Arctic and Antarctic regions. Environ. Sci. Technol. 51, 8944-8952. https://doi.org/10.1021/acs.est.7b02481.

Binnington, M.J., Curren, M.S., Chan, H.M., Wania, F., 2016. Balancing the benefits and costs of traditional food substitution by indigenous Arctic women of childbearing age: Impacts on persistent organic pollutant, mercury, and nutrient intakes. Environ. Int. 94, 554-566. https://doi.org/10.1016/j.envint.2016.06.016.

Blais, J.M., Kimpe, L.E., Mcmahon, D., Keatley, B.E., Mallory, M.L., Douglas, M.S.V., et al., 2005. Arctic seabirds transport marine-derived contaminants. Science. 309, 445. https://doi.org/10.1126/science.1112658.

837 Bogdal, C., Schmid, P., Zennegg, M., Anselmetti, F.S., Scheringer, M., Hungerbühler, K., 2009. Blast
838 from the past: melting glaciers as a relevant source for Persistent Organic Pollutants. Environ. Sci.
839 Technol. 43, 8173-8177. https://doi.org/10.1021/es901628x.
840 Bossi, R., Skjøth, C.A., Skov, H., 2013. Three years (2008-2010) of measurements of atmospheric
841 concentrations of organochlorine pesticides (OCPs) at station Nord, North-East Greenland. Environ.
842 Sci.: Processes Impacts. 15, 2213-2219. https://doi.org/10.1039/c3em00304c.
843 Bossi, R., Vorkamp, K., Skov, H., 2016. Concentrations of organochlorine pesticides, polybrominated
844 diphenyl ethers and perfluorinated compounds in the atmosphere of North Greenland. Environ.
845 Pollut. 217, 4-10. https://doi.org/10.1016/j.envpol.2015.12.026.
846 Botterell, Z.L.R., Beaumont, N., Dorrington, T., Steinke, M., Thompson, R.C., Lindeque, P.K., 2019.
847 Bioavailability and effects of microplastics on marine zooplankton: A review. Environ. Pollut. 245,
848 98-110. https://doi.org/10.1016/j.envpol.2018.10.065.
849 Braune, B.M., 2007. Temporal trends of organochlorines and mercury in seabird eggs from the Canadian
850 Arctic, 1975-2003. Environ. Pollut. 148, 599-613. https://doi.org/10.1016/j.envpol.2006.11.024.
851 Bustnes, J.O., Borgå, K., Dempster, T., Lie, E., Nygård, T., Uglem, I., 2012. Latitudinal distribution of
852 Persistent Organic Pollutants in pelagic and demersal marine fish on the Norwegian coast. Environ.
853 Sci. Technol. 46, 7836-7843. https://doi.org/10.1021/es301191t.
854 Cabrerizo, A., Larramendi, R., Albar, J.-P., Dachs, J., 2017. Persistent organic pollutants in the
855 atmosphere of the Antarctic Plateau. Atmos. Environ. 149, 104-108.
856 https://doi.org/10.1016/j.atmosenv.2016.11.015.
857 Cabrerizo, A., Muir, D.C.G., De Silva, A.O., Wang, X., Lamoureux, S.F., Lafrenière, M.J., 2018a.
858 Legacy and emerging Persistent Organic Pollutants (POPs) in terrestrial compartments in the high
859 Arctic: Sorption and secondary sources. Environ. Sci. Technol. 52, 14187-14197.
860 https://doi.org/10.1021/acs.est.8b05011.

Cabrerizo, A., Muir, D.C.G., Köck, G., Iqaluk, D., Wang, X., 2018b. Climatic influence on temporal trends of polychlorinated biphenyls and organochlorine pesticides in landlocked Arctic char from lakes in the Canadian high Arctic. Environ. Sci. Technol. 52, 10380-10390. https://doi.org/10.1021/acs.est.8b01860

Cabrerizo, A., Muir, D.C.G., Teixeira, C., Lamoureux, S.F., Lafreniere, M.J., 2019. Snow deposition and melting as drivers of Polychlorinated Biphenyls and Organochlorine Pesticides in Arctic rivers, lakes, and ocean. Environ. Sci. Technol. 53, 14377-14386. https://doi.org/10.1021/acs.est.9b05150.

Cai, M., Qiu, C., Shen, Y., Cai, M., Huang, S., Qian, B., et al., 2010. Concentration and distribution of 17 organochlorine pesticides (OCPs) in seawater from the Japan Sea northward to the Arctic Ocean. Sci. China: Chem. 53, 1033-1047. https://doi.org/10.1007/s11426-010-0182-0.

Carlsson, P., Breivik, K., Brorstrom-Lunden, E., Cousins, I., Christensen, J., Grimalt, J.O., et al., 2018. Polychlorinated biphenyls (PCBs) as sentinels for the elucidation of Arctic environmental change processes: a comprehensive review combined with ArcRisk project results. Environ. Sci. Pollut. Res. 25, 22499-22528. https://doi.org/10.1007/s11356-018-2625-7.

Casal, P., Castro-Jimenez, J., Pizarro, M., Katsoyiannis, A., Dachs, J., 2018. Seasonal soil/snow-air exchange of semivolatile organic pollutants at a coastal arctic site (Tromso, 69°N). Sci. Total Environ. 636, 1109-1116. https://doi.org/10.1016/j.scitotenv.2018.04.330.

Chernyak, S.M., McConnell, L.L., Riee, C.P., 1995. Fate of some chlorinated hydrocarbons in Arctic and far eastern ecosystems in the Russian Federation. Sci. Total Environ. 160-161, 75-85. https://doi.org/10.1016/0048-9697(95)04346-3.

Choy, E.S., Kimpe, L.E., Mallory, M.L., Smol, J.P., Blais, J.M., 2010. Contamination of an Arctic terrestrial food web with marine-derived persistent organic pollutants transported by breeding seabirds. Environ. Pollut. 158, 3431-3438. https://doi.org/10.1016/j.envpol.2010.07.014.

884 Corsolini, S., Borghesi, N., Ademollo, N., Focardi, S., 2011. Chlorinated biphenyls and pesticides in
885 migrating and resident seabirds from East and West Antarctica. Environ. Int. 37, 1329-1335.
886 https://doi.org/10.1016/j.envint.2011.05.017.
887 Corsolini, S., Sarà, G., 2017. The trophic transfer of persistent pollutants (HCB, DDTs, PCBs) within
888 polar marine food webs. Chemosphere. 177, 189-199.
889 https://doi.org/10.1016/j.chemosphere.2017.02.116.
890 Cui, S., Zhang, Z., Fu, Q., Hough, R., Yates, K., Osprey, M., et al., 2020. Long-term spatial and temporal
891 patterns of polycyclic aromatic hydrocarbons (PAHs) in Scottish soils over 20 years (1990-2009): A
892 national picture. Geoderma. 361, 114135. https://doi.org/10.1016/j.geoderma.2019.114135.
893 Darby, D.A., Ortiz, J., Polyak, L., Lund, S., Jakobsson, M., Woodgate, R.A., 2009. The role of currents
894 and sea ice in both slowly deposited central Arctic and rapidly deposited Chukchi-Alaskan margin
895 sediments. Global Planet. Change. 68, 58-72. https://doi.org/10.1016/j.gloplacha.2009.02.007.
896 Deutch, B., Pedersen, H.S., Asmund, G., Hansen, J.C., 2007. Contaminants, diet, plasma fatty acids and
897 smoking in Greenland 1999-2005. Sci. Total Environ. 372, 486-496.
898 https://doi.org/10.1016/j.scitotenv.2006.10.043.
899 Ding, X., Wang, X., Wang, Q., Xie, Z., Xiang, C., Mai, B., et al., 2009. Atmospheric DDTs over the
900 North Pacific Ocean and the adjacent Arctic region: Spatial distribution, congener patterns and
901 source implication. Atmos. Environ. 43, 4319-4326.
902 http://dx.doi.org/10.1016/j.atmosenv.2009.06.003.
903 Eckbo, N., Le Bohec, C., Planas-Bielsa, V., Warner, N.A., Schull, Q., Herzke, D., et al., 2019. Individual
904 variability in contaminants and physiological status in a resident Arctic seabird species. Environ.
905 Pollut. 249, 191-199. https://doi.org/10.1016/j.envpol.2019.01.025.
906 Evenset, A., Christensen, G.N., Skotvold, T., Fjeld, E., Schlabach, M., Wartena, E., et al., 2004. A
907 comparison of organic contaminants in two high Arctic lake ecosystems, Bjørnøya (Bear Island),
908 Norway. Sci. Total Environ. 318, 125-141. https://doi.org/10.1016/S0048-9697(03)00365-6.

909 Evenset, A., Hallanger, I.G., Tessmann, M., Warner, N., Ruus, A., Borgå, K., et al., 2016. Seasonal
910 variation in accumulation of persistent organic pollutants in an Arctic marine benthic food web. Sci.
911 Total Environ. 542, 108-120. https://doi.org/10.1016/j.scitotenv.2015.10.092.
912 Ewald, G., Larsson, P., Linge, H., Okla, L., Szarzi, N., 1998. Biotransport of organic pollutants to an
913 inland Alaska lake by migrating Sockeye salmon (*Oncorhyncus nerka*). Arctic. 51, 40-47.
914 https://journalhosting.ucalgary.ca/index.php/arctic/article/download/64099/48034.
915 Fisk, A.T., Hobson, K.A., Norstrom, R.J., 2001. Influence of chemical and biological factors on trophic
916 transfer of Persistent Organic Pollutants in the Northwater Polynya marine food web. Environ. Sci.
917 Technol. 35, 1700-1700. https://doi.org/10.1021/es010719m.
918 Frossard, V., Vagnon, C., Cottin, N., Pin, M., Santoul, F., Naffrechoux, E., 2023. The biological invasion
919 of an apex predator (*Silurus glanis*) amplifies PCB transfer in a large lake food web. Sci. Total
920 Environ. 902, 166037. https://doi.org/10.1016/j.scitotenv.2023.166037.
921 Gao, X., Huang, C., Rao, K., Xu, Y., Huang, Q., Wang, F., et al., 2018. Occurrences, sources, and
922 transport of hydrophobic organic contaminants in the waters of Fildes Peninsula, Antarctica.
923 Environ. Pollut. 241, 950-958. https://doi.org/10.1016/j.envpol.2018.06.025.
924 Gao, X., Huang, P., Huang, Q., Rao, K., Lu, Z., Xu, Y., et al., 2019. Organophosphorus flame retardants
925 and persistent, bioaccumulative, and toxic contaminants in Arctic seawaters: On-board passive
926 sampling coupled with target and non-target analysis. Environ. Pollut. 253, 1-10.
927 https://doi.org/10.1016/j.envpol.2019.06.094.
928 Garmash, O., Hermanson, M.H., Isaksson, E., Schwikowski, M., Divine, D., Teixeira, C., et al., 2013.
929 Deposition history of Polychlorinated Biphenyls to the Lomonosovfonna glacier, Svalbard: A 209
930 congener analysis. Environ. Sci. Technol. 47, 12064-12072. https://doi.org/10.1021/es402430t.
931 Gaudet, C., Lingard, S., Cureton, P., Keenleyside, K., Smith, S., Raju, G., 1995. Canadian Environmental
932 Quality Guidelines for mercury. Water, Air, Soil Pollut. 80, 1149-1159.
933 https://doi.org/10.1007/BF01189777.

Gebbink, W.A., Sonne, C., Dietz, R., Kirkegaard, M., Riget, F.F., Born, E.W., et al., 2008. Tissue-specific congener composition of organohalogen and metabolite contaminants in East Greenland polar bears (*Ursus maritimus*). Environ. Pollut. 152, 621-629. https://doi.org/10.1016/j.envpol.2007.07.001.

Goswami, P., Selvakumar, N., Verma, P., Saha, M., Suneel, V., Vinithkumar, N.V., et al., 2023. Microplastic intrusion into the zooplankton, the base of the marine food chain: Evidence from the Arabian Sea, Indian Ocean. Sci. Total Environ. 864, 160876. https://doi.org/10.1016/j.scitotenv.2022.160876.

Gouin, T., Mackay, D., Jones, K.C., Harner, T., Meijer, S.N., 2004. Evidence for the "grasshopper" effect and fractionation during long-range atmospheric transport of organic contaminants. Environ. Pollut. 128, 139-148. https://doi.org/10.1016/j.envpol.2003.08.025.

Hallanger, I.G., Ruus, A., Warner, N.A., Herzke, D., Evenset, A., Schøyen, M., et al., 2011. Differences between Arctic and Atlantic fjord systems on bioaccumulation of persistent organic pollutants in zooplankton from Svalbard. Sci. Total Environ. 409, 2783-2795. https://doi.org/10.1016/j.scitotenv.2011.03.015.

Hao, Y., Li, Y., Han, X., Wang, T., Yang, R., Wang, P., et al., 2019. Air monitoring of polychlorinated biphenyls, polybrominated diphenyl ethers and organochlorine pesticides in west Antarctica during 2011-2017: Concentrations, temporal trends and potential sources. Environ. Pollut. 249, 381-389. https://doi.org/10.1016/j.envpol.2019.03.039.

He, S., 2013. Chemical characteristic of organochlorine pesticides of marine aerosols collected on the route of Chinese national Antarctic research expedition (In Chinese). Master, Third institute of Oceanorgraphy, Minsitry of Natural Resources, Xiamen.

Helgason, L.B., Arukwe, A., Gabrielsen, G.W., Harju, M., Hegseth, M.N., Heimstad, E.S., et al., 2010. Biotransformation of PCBs in Arctic seabirds: Characterization of phase I and II pathways at

transcriptional, translational and activity levels. Comp. Biochem. Physiol., Part C: Toxicol. Pharmacol. 152, 34-41. https://doi.org/10.1016/j.cbpc.2010.02.009.

Herbert, B.M.J., Halsall, C.J., Villa, S., Fitzpatrick, L., Jones, K.C., Lee, R.G.M., et al., 2005. Polychlorinated naphthalenes in air and snow in the Norwegian Arctic: a local source or an Eastern Arctic phenomenon? Sci. Total Environ. 342, 145-160. https://doi.org/10.1016/j.scitotenv.2004.12.029.

Hobson, K.A., Fisk, A., Karnovsky, N., Holst, M., Gagnon, J.-M., Fortier, M., 2002. A stable isotope (δ^{13}C, δ^{15}N) model for the North Water food web: implications for evaluating trophodynamics and the flow of energy and contaminants. Deep Sea Res., Part II. 49, 5131-5150. https://doi.org/10.1016/S0967-0645(02)00182-0.

Huang, Y., Yue, X., Li, J., Xu, W., Zhang, G., Cheng, Z., et al., 2013. Organochlorine Pesticides in the atmosphere and surface water from the equatorial Indian Ocean: Enantiomeric signatures, sources, and fate. Environ. Sci. Technol. 47, 13395-13403. https://doi.org/10.1021/es403138p.

Hung, H., Blanchard P., Halsall C.J., Bidleman T.F., Stern G.A., Fellin P., et al., 2005. Temporal and spatial variabilities of atmospheric polychlorinated biphenyls (PCBs), organochlorine (OC) pesticides and polycyclic aromatic hydrocarbons (PAHs) in the Canadian Arctic: Results from a decade of monitoring. Sci. Total Environ. 342, 119-144. https://doi.org/10.1016/j.scitotenv.2004.12.058.

Hung, H., Halsall, C.J., Blanchard, P., Li, H.H., Fellin, P., Stern, G., et al., 2001. Are PCBs in the Canadian Arctic atmosphere declining? Evidence from 5 years of monitoring. Environ. Sci. Technol. 35, 1303-1311. https://doi.org/10.1021/es001704b.

Hung, H., Kallenborn, R., Breivik, K., Su, Y., Brorström-Lundén, E., Olafsdottir, K., et al., 2010. Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993-2006. Sci. Total Environ. 408, 2854-2873. https://doi.org/10.1016/j.scitotenv.2009.10.044.

Hung, H., Katsoyiannis, A.A., Brorström-Lundén, E., Olafsdottir, K., Aas, W., Breivik, K., et al., 2016. Temporal trends of Persistent Organic Pollutants (POPs) in arctic air: 20 years of monitoring under the Arctic Monitoring and Assessment Programme (AMAP). Environ. Pollut. 217, 52-61. https://doi.org/10.1016/j.envpol.2016.01.079.

Iversen, T., 1996. Atmospheric transport pathways for the Arctic, Berlin, Heidelberg.

Jantunen, L.M., Wong, F., Gawor, A., Kylin, H., Helm, P.A., Stern, G.A., et al., 2015. 20 years of air-water gas exchange observations for pesticides in the western Arctic Ocean. Environ. Sci. Technol. 49, 13844-13852. https://doi.org/10.1021/acs.est.5b01303.

Jartun, M., Ottesen, R.T., Volden, T., Lundkvist, Q., 2009. Local sources of polychlorinated biphenyls (PCB) in Russian and Norwegian settlements on Spitsbergen Island, Norway. J. Toxicol. Environ. Health, Part A. 72, 284-294. https://doi.org/10.1080/15287390802539426.

Jaspers, V.L.B., Sonne, C., Soler-Rodriguez, F., Boertmann, D., Dietz, R., Eens, M., et al., 2013. Persistent organic pollutants and methoxylated polybrominated diphenyl ethers in different tissues of white-tailed eagles (*Haliaeetus albicilla*) from West Greenland. Environ. Pollut. 175, 137-146. https://doi.org/10.1016/j.envpol.2012.12.023.

Jhamtani, R.C., Shukla, S., Sivaperumal, P., Dahiya, M.S., Agarwal, R., 2018. Impact of co-exposure of aldrin and titanium dioxide nanoparticles at biochemical and molecular levels in Zebrafish. Environ. Toxicol. Pharmacol. 58, 141-155. https://doi.org/10.1016/j.etap.2017.12.021.

Johansen, S., Poste, A., Allan, I., Evenset, A., Carlsson, P., 2021. Terrestrial inputs govern spatial distribution of polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB) in an Arctic fjord system (Isfjorden, Svalbard). Environ. Pollut. 281, 116963. https://doi.org/10.1016/j.envpol.2021.116963.

Jones, K.C., Halsall, C.J., Herbert, B., Kallenborn, R., Villa, S., 2005. Rapid changes in PCB and OC Pesticide concentrations in Arctic snow. Environ. Sci. Technol. 39, 2998-3005. https://doi.org/10.1021/es040076l.

1008 Kahkashan, S., Wang, X., Chen, J., Bai, Y., Ya, M., Wu, Y., et al., 2019. Concentration, distribution and
1009 sources of perfluoroalkyl substances and organochlorine pesticides in surface sediments of the
1010 northern Bering Sea, Chukchi Sea and adjacent Arctic Ocean. Chemosphere. 235, 959-968.
1011 https://doi.org/10.1016/j.chemosphere.2019.06.219.
1012 Kallenborn, R., Halsall, C., Dellong, M., Carlsson, P., 2012. The influence of climate change on the
1013 global distribution and fate processes of anthropogenic persistent organic pollutants. J. Environ.
1014 Monit. 14, 2854-2869. https://doi.org/10.1039/C2EM30519D.
1015 Kallenborn, R., Ottesen, R.T., Gabrielsen, G.W., Schrum, C., Evenset, A., Ruus, A., et al., 2013. PCBs on
1016 Svalbard (Report 2011): Governor of Svalbard. PCBs on Svalbard: Status of knowledge and
1017 management, Svalbard.
1018 Kelly, B.C., Ikonomou, M.G., Blair, J.D., Gobas, F.A.P.C., 2008. Bioaccumulation behaviour of
1019 polybrominated diphenyl ethers (PBDEs) in a Canadian Arctic marine food web. Sci. Total Environ.
1020 401, 60-72. https://doi.org/10.1016/j.scitotenv.2008.03.045.
1021 Kelly, B.C., Ikonomou, M.G., Blair, J.D., Surridge, B., Hoover, D., Grace, R., et al., 2009. Perfluoroalkyl
1022 contaminants in an Arctic marine food web: Trophic magnification and wildlife exposure. Environ.
1023 Sci. Technol. 43, 4037-4043. https://doi.org/10.1021/es9003894.
1024 Khairy, M.A., Luek, J.L., Dickhut, R., Lohmann, R., 2016. Levels, sources and chemical fate of persistent
1025 organic pollutants in the atmosphere and snow along the western Antarctic Peninsula. Environ.
1026 Pollut. 216, 304-313. https://doi.org/10.1016/j.envpol.2016.05.092.
1027 Kim, J.T., Choi, Y.J., Barghi, M., Kim, J.H., Jung, J.W., Kim, K., et al., 2021. Occurrence, distribution,
1028 and bioaccumulation of new and legacy persistent organic pollutants in an ecosystem on King
1029 George Island, maritime Antarctica. J. Hazard. Mater. 405, 124141.
1030 https://doi.org/10.1016/j.jhazmat.2020.124141.

Kodavanti, P.R.S., Curras-Collazo, M.C., 2010. Neuroendocrine actions of organohalogens: thyroid hormones, arginine vasopressin, and neuroplasticity. Front. Neuroendocrinol. 31, 479-496. https://doi.org/10.1016/j.yfrne.2010.06.005.

Lakhmanov, D., Varakina, Y., Aksenov, A., Sorokina, T., Sobolev, N., Kotsur, D., et al., 2020. Persistent Organic Pollutants (POPs) in fish consumed by the indigenous peoples from Nenets Autonomous Okrug. Environments. 7, 3. https://doi.org/10.3390/environments7010003.

Lebrun, J.D., Geffard, O., Urien, N., François, A., Uher, E., Fechner, L.C., 2015. Seasonal variability and inter-species comparison of metal bioaccumulation in caged gammarids under urban diffuse contamination gradient: Implications for biomonitoring investigations. Sci. Total Environ. 511, 501-508. https://doi.org/10.1016/j.scitotenv.2014.12.078.

Li, Y., Geng, D., Liu, F., Wang, T., Wang, P., Zhang, Q., et al., 2012. Study of PCBs and PBDEs in King George Island, Antarctica, using PUF passive air sampling. Atmos. Environ. 51, 140-145. https://doi.org/10.1016/j.atmosenv.2012.01.034.

Li, Y.F., Macdonald, R.W., 2005. Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review. Sci. Total Environ. 342, 87-106. https://doi.org/10.1016/j.scitotenv.2004.12.027.

Lie, E., Bernhoft, A., Riget, F., Belikov, S.E., Boltunov, A.N., Derocher, A.E., et al., 2003. Geographical distribution of organochlorine pesticides (OCPs) in polar bears (*Ursus maritimus*) in the Norwegian and Russian Arctic. Sci. Total Environ. 306, 159-170. https://doi.org/10.1016/S0048-9697(02)00490-4.

Lin, T., Hu, L., Shi, X., Li, Y., Guo, Z., Zhang, G., 2012. Distribution and sources of organochlorine pesticides in sediments of the coastal East China Sea. Mar. Pollut. Bull. 64, 1549-1555. https://doi.org/10.1016/j.marpolbul.2012.05.021.

Lomartire, S., Marques, J.C., Gonçalves, A.M.M., 2021. The key role of zooplankton in ecosystem services: A perspective of interaction between zooplankton and fish recruitment. Ecol. Indic. 129, 107867. https://doi.org/10.1016/j.ecolind.2021.107867.

Lu, Q., Jürgens, M.D., Johnson, A.C., Graf, C., Sweetman, A., Crosse, J., et al., 2017. Persistent Organic Pollutants in sediment and fish in the River Thames Catchment (UK). Sci. Total Environ. 576, 78-84. https://doi.org/10.1016/j.scitotenv.2016.10.067.

Luttmer, C., Ficko, S., Reimer, K., Zeeb, B., 2013. Deciduous vegetation (*Betula glandulosa*) as a biomonitor of airborne PCB contamination from a local source in the Arctic. Sci. Total Environ. 445-446, 314-320. https://doi.org/10.1016/j.scitotenv.2012.12.063.

Ma, J., Hung, H., Tian, C., Kallenborn, R., 2011. Revolatitization of persistent organic pollutants in the Arctic induced by climate change. Nat. Clim. Change. 1, 255-260. https://doi.org/10.1038/nclimate1167.

Ma, Y., Xie, Z., Halsall, C., Möller, A., Yang, H., Zhong, G., et al., 2015. The spatial distribution of organochlorine pesticides and halogenated flame retardants in the surface sediments of an Arctic fjord: The influence of ocean currents vs. glacial runoff. Chemosphere. 119, 953-960. https://doi.org/10.1016/j.chemosphere.2014.09.012.

Macdonald, R.W., Barrie, L.A., Bidleman, T.F., Diamond, M.L., Gregor, D.J., Semkin, R.G., et al., 2000. Contaminants in the Canadian Arctic: 5 years of progress in understanding sources, occurrence and pathways. Sci. Total Environ. 254, 93-234. https://doi.org/10.1016/S0048-9697(00)00434-4.

Macdonald, R.W., Harner, T., Fyfe, J., 2005. Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. Sci. Total Environ. 342, 5-86. https://doi.org/10.1016/j.scitotenv.2004.12.059.

Malanichev, A., Mantseva, E., Shatalov, V., Strukov, B., Vulykh, N., 2004. Numerical evaluation of the PCBs transport over the northern hemisphere. Environ. Pollut. 128, 279-289. https://doi.org/10.1016/j.envpol.2003.08.040.

1079 Mandalakis, M., Berresheim, H., Stephanou, E.G., 2003. Direct evidence for destruction of
1080 Polychlorobiphenyls by OH radicals in the subtropical troposphere. Environ. Sci. Technol. 37, 542-
1081 547. https://doi.org/10.1021/es020163i.
1082 Masset, T., Frossard, V., Perga, M.E., Cottin, N., Piot, C., Cachera, S., et al., 2019. Trophic position and
1083 individual feeding habits as drivers of differential PCB bioaccumulation in fish populations. Sci.
1084 Total Environ. 674, 472-481. https://doi.org/10.1016/j.scitotenv.2019.04.196.
1085 Matthies, M., Klasmeier, J., Beyer, A., Ehling, C., 2009. Assessing persistence and long-range transport
1086 potential of current-use pesticides. Environ. Sci. Technol. 43, 9223-9229.
1087 https://doi.org/10.1021/es900773u.
1088 McGovern, M., Warner, N.A., Borgå, K., Evenset, A., Carlsson, P., Skogsberg, E., et al., 2022. Is glacial
1089 meltwater a secondary source of legacy contaminants to Arctic coastal food webs? Environ. Sci.
1090 Technol. 56, 6337-6348. 10.1021/acs.est.1c07062.
1091 Meerts, I.A.T.M., Yvonne, A., Cenijn, P.H., van den Berg, J.H.J., Weijers, B.M., Bergman, A., et al.,
1092 2002. Placental transfer of a hydroxylated polychlorinated biphenyl and effects on fetal and
1093 maternal thyroid hormone homeostasis in the rat. Toxicol. Sci. 68, 361-371.
1094 https://doi.org/10.1093/toxsci/68.2.361.
1095 Michelutti, N., Liu, H., Smol, J.P., Kimpe, L.E., Keatley, B.E., Mallory, M., et al., 2009. Accelerated
1096 delivery of polychlorinated biphenyls (PCBs) in recent sediments near a large seabird colony in
1097 Arctic Canada. Environ. Pollut. 157, 2769-2775. https://doi.org/10.1016/j.envpol.2009.04.025.
1098 Miljeteig, C., Gabrielsen, G.W., Strom, H., Gavrilo, M.V., Lie, E., Jenssen, B.M., 2012. Eggshell
1099 thinning and decreased concentrations of vitamin E are associated with contaminants in eggs of
1100 ivory gulls. Sci. Total Environ. 431, 92-99. https://doi.org/10.1016/j.scitotenv.2012.05.018.
1101 Miljeteig, C., Strøm, H., Gavrilo, M.V., Volkov, A., Jenssen, B.M., Gabrielsen, G.W., 2009. High levels
1102 of contaminants in Ivory Gull *Pagophila eburnea* eggs from the Russian and Norwegian Arctic.
1103 Environ. Sci. Technol. 43, 5521-5528. https://doi.org/10.1021/es900490n.

Montone, R.C., Taniguchi, S., Weber, R.R., 2003. PCBs in the atmosphere of King George Island, Antarctica. Sci. Total Environ. 308, 167-173. https://doi.org/10.1016/S0048-9697(02)00649-6.

Moreira, B.P., Silva, J.F., Jarak, I., de Lourdes Pereira, M., Oliveira, P.F., Alves, M.G., 2020. Technical-grade chlordane compromises rat Sertoli cells proliferation, viability and metabolic activity. Toxicol. In Vitro. 63, 104673. https://doi.org/10.1016/j.tiv.2019.104673.

Morris, A.D., Muir, D.C.G., Solomon, K.R., Letcher, R.J., McKinney, M.A., Fisk, A.T., et al., 2016. Current-use pesticides in seawater and their bioaccumulation in polar bear-ringed seal food chains of the Canadian Arctic. Environ. Toxicol. Chem. 35, 1695-1707. https://doi.org/10.1002/etc.3427.

Moses, S.K., Harley, J.R., Lieske, C.L., Muir, D.C.G., Whiting, A.V., O'Hara, T.M., 2015. Variation in bioaccumulation of persistent organic pollutants based on octanol-air partitioning: Influence of respiratory elimination in marine species. Mar. Pollut. Bull. 100, 122-127. https://doi.org/10.1016/j.marpolbul.2015.09.020.

Muir, D., Savinova, T., Savinov, V., Alexeeva, L., Potelov, V., Svetochev, V., 2003. Bioaccumulation of PCBs and chlorinated pesticides in seals, fishes and invertebrates from the White Sea, Russia. Sci. Total Environ. 306, 111-131. https://doi.org/10.1016/S0048-9697(02)00488-6.

Nash, S.M.B., Wild, S.J., Hawker, D.W., Cropp, R.A., Hung, H., Wania, F., et al., 2017. Persistent Organic Pollutants in the east Antarctic atmosphere: Inter-annual observations from 2010-2015 using high-flow-through passive sampling. Environ. Sci. Technol. 51, 13929-13937. https://doi.org/10.1021/acs.est.7b04224.

Neerland, E.D., Bytingsvik, J., Nikiforov, V.A., Evenset, A., Åse, K., 2019. DNA Double-Strand Breaks in Arctic Char (*Salvelinus alpinus*) from Bjørnøya in the Norwegian Arctic. Environ. Toxicol. Chem. 38, 2405-2413. https://doi.org/10.1002/etc.4546.

Nfon, E., Cousins, I.T., 2007. Modelling PCB bioaccumulation in a Baltic food web. Environ. Pollut. 148, 73-82. https://doi.org/10.1016/j.envpol.2006.11.033.

Niknam, Y., Feng, W., Cherednichenko, G., Dong, Y., Joshi, S.N., Vyas, S.M., et al., 2013. Structure-activity relationship of selected meta- and para-hydroxylated non-dioxin like polychlorinated biphenyls: from single RyR1 channels to muscle dysfunction. Toxicol. Sci. 136, 500-513. https://doi.org/10.1093/toxsci/kft202.

Nost, T.H., Helgason, L.B., Harju, M., Heimstad, E.S., Gabrielsen, G.W., Jenssen, B.M., 2012. Halogenated organic contaminants and their correlations with circulating thyroid hormones in developing Arctic seabirds. Sci. Total Environ. 414, 248-256. https://doi.org/10.1016/j.scitotenv.2011.11.051.

Nuijten, R.J.M., Hendriks, A.J., Jenssen, B.M., Schipper, A.M., 2016. Circumpolar contaminant concentrations in polar bears (*Ursus maritimus*) and potential population-level effects. Environ. Res. 151, 50-57. https://doi.org/10.1016/j.envres.2016.07.021.

Pacyna-Kuchta, A.D., Wietrzyk-Pelka, P., Wegrzyn, M.H., Frankowski, M., Polkowska, Z., 2020. A screening of select toxic and essential elements and persistent organic pollutants in the fur of Svalbard reindeer. Chemosphere. 245, 125458. https://doi.org/10.1016/j.chemosphere.2019.125458.

Palm, A., Cousins, I., Gustafsson, Ö., Axelman, J., Grunder, K., Broman, D., et al., 2004. Evaluation of sequentially-coupled POP fluxes estimated from simultaneous measurements in multiple compartments of an air-water-sediment system. Environ. Pollut. 128, 85-97. https://doi.org/10.1016/j.envpol.2003.08.023.

Pedro, S., Boba, C., Dietz, R., Sonne, C., Rosing-Asvid, A., Hansen, M., et al., 2017. Blubber-depth distribution and bioaccumulation of PCBs and organochlorine pesticides in Arctic-invading killer whales. Sci. Total Environ. 601-602, 237-246. https://doi.org/10.1016/j.scitotenv.2017.05.193.

Pozo, K., Harner, T., Wania, F., Muir, D.C.G., Jones, K.C., Barrie, L.A., 2006. Toward a global network for persistent organic pollutants in air: results from the GAPS study. Environ. Sci. Technol. 40, 4867-4873. https://doi.org/10.1021/es060447t.

Pućko, M., Stern, G.A., Burt, A.E., Jantunen, L.M., Bidleman, T.F., Macdonald, R.W., et al., 2017. Current use pesticide and legacy organochlorine pesticide dynamics at the ocean-sea ice-atmosphere interface in resolute passage, Canadian Arctic, during winter-summer transition. Sci. Total Environ. 580, 1460-1469. https://doi.org/10.1016/j.scitotenv.2016.12.122.

Pućko, M., Stern, G.A., Macdonald, R.W., Barber, D.G., Rosenberg, B., Walkusz, W., 2013. When will α-HCH disappear from the western Arctic Ocean? J. Marine Syst. 27, 88-100. https://doi.org/10.1016/j.jmarsys.2011.09.007.

Purnomo, A.S., Mori, T., Putra, S.R., Kondo, R., 2013. Biotransformation of heptachlor and heptachlor epoxide by white-rot fungus *Pleurotus ostreatus*. Int. Biodeterior. Biodegrad. 82, 40-44. https://doi.org/10.1016/j.ibiod.2013.02.013.

Qiu, X., Zhu, T., Li, J., Pan, H., Li, Q., Miao, G., et al., 2004. Organochlorine Pesticides in the air around the Taihu Lake, China. Environ. Sci. Technol. 38, 1368-1374. https://doi.org/10.1021/es035052d.

Rahman, M.M., Lee, H.S., Abd El-Aty, A.M., Kabir, M.H., Chung, H.S., Park, J.-H., et al., 2018. Determination of endrin and δ-keto endrin in five food products of animal origin using GC-μECD: A modified QuEChERS approach to traditional detection. Food Chem. 263, 59-66. https://doi.org/10.1016/j.foodchem.2018.04.099.

Routti, H., Andersen, M.S., Fuglei, E., Polder, A., Yoccoz, N.G., 2016. Concentrations and patterns of hydroxylated polybrominated diphenyl ethers and polychlorinated biphenyls in arctic foxes (*Vulpes lagopus*) from Svalbard. Environ. Pollut. 216, 264-272. https://doi.org/10.1016/j.envpol.2016.05.056.

Scheringer, M., Jones, K.C., Matthies, M., Simonich, S., van de Meent, D., 2009. Multimedia partitioning, overall persistence, and long-range transport potential in the context of POPs and PBT chemical assessments. Integr. Environ. Assess. Manag. 5, 557-576. https://doi.org/10.1897/ieam_2009-007.1.

Sobek, A., Mclachlan, M.S., Borga, K., Asplund, L., Lundstedt-Enkel, K., Polder, A., et al., 2010. A comparison of PCB bioaccumulation factors between an arctic and a temperate marine food web. Sci. Total Environ. 408, 2753-2760. https://doi.org/10.1016/j.scitotenv.2010.03.013.

Sonne, C., 2010. Health effects from long-range transported contaminants in Arctic top predators: An integrated review based on studies of polar bears and relevant model species. Environ. Int. 36, 461-491. https://doi.org/10.1016/j.envint.2010.03.002.

Stern, A.H., 2014. Hazard identification of the potential for dieldrin carcinogenicity to humans. Environ. Res. 131, 188-214. https://doi.org/10.1016/j.envres.2014.02.007.

Stocker, J., Scheringer, M., Wegmann, F., Hungerbühler, K., 2007. Modeling the effect of snow and ice on the global environmental fate and long-range transport potential of semivolatile organic compounds. Environ. Sci. Technol. 41, 6192-6198. https://doi.org/10.1021/es062873k.

Strid, A., Jörundsdóttir, H., Päpke, O., Svavarsson, J., Bergman, Å., 2007. Dioxins and PCBs in Greenland shark (*Somniosus microcephalus*) from the North-East Atlantic. Mar. Pollut. Bull. 54, 1514-1522. https://doi.org/10.1016/j.marpolbul.2007.04.018.

Su, Y., Hung, H., Blanchard, P., Patton, G.W., Kallenborn, R., Konoplev, A., et al., 2006. Spatial and seasonal variations of Hexachlorocyclohexanes (HCHs) and Hexachlorobenzene (HCB) in the Arctic atmosphere. Environ. Sci. Technol. 40, 6601-6607. https://doi.org/10.1021/es061065q.

Su, Y., Hung, H., Blanchard, P., Patton, G.W., Kallenborn, R., Konoplev, A., et al., 2008. A circumpolar perspective of atmospheric organochlorine pesticides (OCPs): Results from six Arctic monitoring stations in 2000-2003. Atmos. Environ. 42, 4682-4698. https://doi.org/10.1016/j.atmosenv.2008.01.054.

Szczybelski, A.S., van den Heuvel-Greve, M.J., Kampen, T., Wang, C., van den Brink, N.W., Koelmans, A.A., 2016. Bioaccumulation of polycyclic aromatic hydrocarbons, polychlorinated biphenyls and hexachlorobenzene by three Arctic benthic species from Kongsfjorden (Svalbard, Norway). Mar. Pollut. Bull. 112, 65-74. https://doi.org/10.1016/j.marpolbul.2016.08.041.

Tartu, S., Fisk, A.T., Götsch, A., Kovacs, K.M., Lydersen, C., Routti, H., 2020. First assessment of pollutant exposure in two balaenopterid whale populations sampled in the Svalbard Archipelago, Norway. Sci. Total Environ. 718, 137327. https://doi.org/10.1016/j.scitotenv.2020.137327.

Ubl, S., Scheringer, M., Stohl, A., Burkhart, J.F., Hungerbuhler, K., 2012. Primary source regions of polychlorinated biphenyls (PCBs) measured in the Arctic. Atmos. Environ. 62, 391-399. https://doi.org/10.1016/j.atmosenv.2012.07.061.

Vecchiato, M., Argiriadis, E., Zambon, S., Barbante, C., Toscano, G., Gambaro, A., et al., 2015. Persistent Organic Pollutants (POPs) in Antarctica: Occurrence in continental and coastal surface snow. Microchem. J. 119, 75-82. https://doi.org/10.1016/j.microc.2014.10.010.

Verhaert, V., Newmark, N., D'Hollander, W., Covaci, A., Vlok, W., Wepener, V., et al., 2017. Persistent organic pollutants in the Olifants River Basin, South Africa: Bioaccumulation and trophic transfer through a subtropical aquatic food web. Sci. Total Environ. 586, 792-806. https://doi.org/10.1016/j.scitotenv.2017.02.057.

Verreault, J., Muir, D.C.G., Norstrom, R.J., Stirling, I., Fisk, A.T., Gabrielsen, G.W., et al., 2005. Chlorinated hydrocarbon contaminants and metabolites in polar bears (*Ursus maritimus*) from Alaska, Canada, East Greenland, and Svalbard: 1996-2002. Sci. Total Environ. 351-352, 369-390. https://doi.org/10.1016/j.scitotenv.2004.10.031.

Verreault, J., Shahmiri, S., Gabrielsen, G.W., Letcher, R.J., 2007. Organohalogen and metabolically-derived contaminants and associations with whole body constituents in Norwegian Arctic glaucous gulls. Environ. Int. 33, 823-830. https://doi.org/10.1016/j.envint.2007.03.013.

Veyrand, B., Venisseau, A., Marchand, P., Antignac, J.-P., Bizec, B.L., 2008. Determination of toxaphene specific congeners in fish liver oil and feeding stuff using gas chromatography coupled to high resolution mass spectrometry. J. Chromatogr. B. 865, 121-126. https://doi.org/10.1016/j.jchromb.2008.02.018.

Vorkamp, K., Balmer, J., Hung, H., Letcher, R.J., Rigét, F.F., 2019. A review of chlorinated paraffin contamination in Arctic ecosystems. Emerging Contam. 5, 219-231. https://doi.org/10.1016/j.emcon.2019.06.001.

Walker, K., Vallero, D.A., Lewis, R.G., 1999. Factors influencing the distribution of Lindane and other Hexachlorocyclohexanes in the environment. Environ. Sci. Technol. 33, 4373-4378. https://doi.org/10.1021/es990647n.

Wang, S., Salamova, A., Hites, R.A., Venier, M., 2018. Spatial and seasonal distributions of Current Use Pesticides (CUPs) in the atmospheric particulate phase in the Great Lakes Region. Environ. Sci. Technol. 52, 6177-6186. https://doi.org/10.1021/acs.est.8b00123.

Wania, F., 2003. Assessing the potential of persistent organic chemicals for long-range transport and accumulation in polar regions. Environ. Sci. Technol. 37, 1344-1351. https://doi.org/10.1021/es026019e.

Wania, F., Dugani, C.B., 2003. Assessing the long-range transport potential of polybrominated diphenyl ethers: a comparison of four multimedia models. Environ. Toxicol. Chem. 22, 1252-1261. https://doi.org/10.1002/etc.5620220610.

Wania, F., Shen, L., Lei, Y., Teixeira, C., Muir, D.C.G., 2003. Development and calibration of a resin-based passive sampling system for monitoring Persistent Organic Pollutants in the atmosphere. Environ. Sci. Technol. 37, 1352-1359. https://doi.org/10.1021/es026166c.

Warner, N.A., Sagerup, K., Kristoffersen, S., Herzke, D., Gabrielsen, G.W., Jenssen, B.M., 2019. Snow buntings (*Plectrophenax nivealis*) as bio-indicators for exposure differences to legacy and emerging persistent organic pollutants from the Arctic terrestrial environment on Svalbard. Sci. Total Environ. 667, 638-647. https://doi.org/10.1016/j.scitotenv.2019.02.351.

Wong, F., Hung, H., Dryfhout-Clark, H., Aas, W., Bohlin-Nizzetto, P., Breivik, K., et al., 2021. Time trends of persistent organic pollutants (POPs) and Chemicals of Emerging Arctic Concern (CEAC)

in Arctic air from 25 years of monitoring. Sci. Total Environ. 775, 145109. https://doi.org/10.1016/j.scitotenv.2021.145109.

Wu, X., Kammerer, A., Lehmler, H.-J., 2014. Microsomal oxidation of 2,2′,3,3′,6,6′-Hexachlorobiphenyl (PCB 136) results in species-dependent chiral signatures of the hydroxylated metabolites. Environ. Sci. Technol. 48, 2436-2444. https://doi.org/10.1021/es405433t.

Wu, X., Lam, J.C.W., Xia, C., Kang, H., Xie, Z., Lam, P.K.S., 2011. Atmospheric concentrations of DDTs and chlordanes measured from Shanghai, China to the Arctic Ocean during the third China Arctic Research Expedition in 2008. Atmos. Environ. 45, 3750-3757. https://doi.org/10.1016/j.atmosenv.2011.04.012.

Xu, X., Wong, C.Y., Tam, N.F.Y., Lo, H.-S., Cheung, S.-G., 2020. Microplastics in invertebrates on soft shores in Hong Kong: Influence of habitat, taxa and feeding mode. Sci. Total Environ. 715, 136999. https://doi.org/10.1016/j.scitotenv.2020.136999.

Ya, M., Wu, Y., Li, Y., Wang, X., 2017. Anthropogenic organochlorine compounds as potential tracers for regional water masses: A case study of estuarine plume, coastal eddy, wind-driven upwelling and long-range warm current. Chemosphere. 170, 75-82. https://doi.org/10.1016/j.chemosphere.2016.12.010.

Yao, Z., Jiang, G., Xu, H., 2002. Distribution of organochlorine pesticides in seawater of the Bering and Chukchi Sea. Environ. Pollut. 116, 49-56. https://doi.org/10.1016/S0269-7491(01)00134-8.

Yu, Y., Hung, H., Alexandrou, N., Roach, P., Nordin, K., 2015. Multiyear measurements of Flame Retardants and Organochlorine Pesticides in air in Canada's Western sub-Arctic. Environ. Sci. Technol. 49, 8623-8630. https://doi.org/10.1021/acs.est.5b01996.

Zhang, F.L., Yang, X.J., Xue, X.L., Tao, X.Q., Lu, G.N., Dang, Z., 2013. Estimation of *n*-octanol/water partition coefficients (log K_{OW}) of polychlorinated biphenyls by using quantum chemical descriptors and partial leasts quares. J. Chem-Ny. 2013, 1-8. https://doi.org/10.1155/2013/740548.

1273 Zhang, P., Ge, L., Gao, H., Yao, T., Fang, X., Zhou, C., et al., 2014. Distribution and transfer pattern of
1274 Polychlorinated Biphenyls (PCBs) among the selected environmental media of Ny-Ålesund, the
1275 Arctic: As a case study. Mar. Pollut. Bull. 89, 267-275.
1276 https://doi.org/10.1016/j.marpolbul.2014.09.050.
1277 Zheng, Y., Han, B., Xu, X., Liu, A., Zheng, L., 2022. Distribution characteristics, source analysis and risk
1278 assessment of organochlorine pesticides in Ny-Ålesund, Arctic. Mar. Pollut. Bull. 181, 113862.
1279 https://doi.org/10.1016/j.marpolbul.2022.113862.
1280 Zhong, G., Xie, Z., Cai, M., Möller, A., Sturm, R., Tang, J., et al., 2012. Distribution and air-sea
1281 exchange of current-use pesticides (CUPs) from East Asia to the high Arctic Ocean. Environ. Sci.
1282 Technol. 46, 259-267. https://doi.org/10.1021/es202655k.
1283 Zhulidov, A.V., Headley, J.V., Pavlov, D.F., Robarts, R.D., Korotova, L.G., Vinnikov, Y.Y., et al., 2000.
1284 Riverine fluxes of the persistent organochlorine pesticides hexachlorcyclohexane and DDT in the
1285 Russian Federation. Chemosphere. 41, 829-841. https://doi.org/10.1016/S0045-6535(99)00520-2.

1286