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Terrestrial permafrost areas – the state of knowledge on transport, fate and degradation of halogenated organic compounds





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The Norwegian Polar Institute is Norway's central governmental institution for management-related research, mapping and environmental monitoring in the Arctic and the Antarctic. The Institute advises Norwegian authorities on matters concerning polar environmental management and is the official environmental management body for Norway's Antarctic territorial claims. The Institute is a Directorate within the Ministry of Climate and Environment.

Norsk Polarinstitutt er Norges hovedinstitusjon for kartlegging, miljøovervåking og forvaltningsrettet forskning i Arktis og Antarktis. Instituttet er faglig og strategisk rådgiver i miljøvernaker i disse områdene og har forvaltningsmyndighet i norsk del av Antarktis. Instituttet er et direktorat under Klima- og miljødepartementet.

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Preface

The Ministry of Climate and Environment has tasked the Norwegian Polar Institute with reporting on the state of knowledge on transport, fate and degradation of priority organic compounds in terrestrial permafrost areas. The report is based on existing literature, monitoring data and results of technical surveys.

The focus here is on halogenated organic contaminants (HOCs) including polychlorinated biphenyls (PCBs), organochlorinated pesticides, brominated flame retardants and perfluoroalkyl and polyfluoroalkyl substances (PFASs). Literature on sources, fate and degradation of HOCs in Arctic permafrost areas is summarized. In addition impact of climate change and implications for Svalbard are shortly discussed.

English summary

Permafrost is ground (soil, rock or sediment) that is 0 °C or colder for at least two consecutive years. It has a widespread distribution in the northern hemisphere with the majority being in the Arctic. Above the permafrost layer is the active layer, which is the layer of ground that thaws during spring and summer and freezes in autumn and winter. The presence and depth of permafrost and the active layer is dependent upon many factors including temperature, topography, vegetation, snow cover, ocean currents and ground material. Due to global climate change permafrost is thawing and permafrost temperatures have generally increased by 0.5-2 °C since the late 1970s. Temperature in the upper part of the permafrost in Svalbard has risen on average 0.7 °C per decade, with an accelerating speed during the past decade. Also, the active layer has become 25–30 cm thicker.

Halogenated organic contaminants (HOCs) including polychlorinated biphenyls (PCBs), organochlorinated pesticides, brominated flame retardants and perfluoroalkyl and polyfluoroalkyl substances (PFASs) are man-made chemicals used for industrial and agricultural purposes. HOCs are transported to pristine Arctic areas through air and ocean currents from more southern regions, but there are also some local sources. Once the contaminants reach the Arctic, their fate depends largely on their physico-chemical properties.

Snow is an efficient scavenger of HOCs and is an important route of wet deposition. Once stored in snowpack, HOCs can be re-volatilized or chemically degraded. The storage capacity of snow is reduced as the snowpack ages, leading to remobilization of contaminants. Therefore, concentrations of HOCs in snow are not constant and the amount of contaminants in meltwater of snow is often less than the amount initially deposited. As glaciers are formed and grow as a result of accumulation of snow, the same processes are applicable to them as well. Contaminant concentrations in soil are generally low except at sites close to local sources. In the archipelago of Svalbard, the highest concentrations of PCBs in soil have been reported from the Russian settlements, where levels are approximately one hundred times higher compared to background levels.

Climate change may lead to increased deposition of HOCs on permafrost land areas as the occurrence of rain over land will increase. This is likely to occur in Svalbard where the winters have become warmer with more precipitation falling as rain. As the temperature of permafrost globally, including Svalbard, is increasing and the depth of the active layer is expanding, the hitherto contained contaminants will be more prone to move from soil and other depository compartments (water and ice) to the air. In addition, warming temperatures may mobilize HOCs from polluted sites more rapidly. The shrinking glaciers will also release stored HOCs, but the extent of this varies.

Norsk sammendrag

Permafrost er grunn (jord, stein eller sediment) som er 0 °C eller kaldere i minst to år på rad. Permafrost har en omfattende distribusjon på den nordlige halvkule og størstedelen er lokalisert i Arktis. Tilstedeværelsen av permafrost er avhengig av flere faktorer som, for eksempel, temperatur, topografi, vegetasjon, snødekke, havstrømmer og grunnmateriale. På grunn av klimaendringene tiner permafrosten, og temperaturen på permafrosten har økt med 0.5-2 °C siden slutten av 1970-tallet. Temperaturen i den øvre delen av permafrosten på Svalbard har i gjennomsnitt steget med 0.7 °C per tiår med akselererende økning det siste tiåret. Det aktive laget ligger over permafrosten og er det laget som tiner om våren og sommeren og fryser igjen om høsten og vinteren. På grunn av at det aktive laget årlig overskrider 0 °C er ikke dette laget en del av permafrosten. Det aktive laget har også blitt 25–30 cm tykkere som følge av de økende temperaturene.

Halogenerte organiske giftstoffer (HOC), inkludert polyklorerte bifenyler (PCB), organoklorerte pesticider, bromerte flammehemmere og perfluoroalkyl stoffer (PFAS), er menneskeskapt kjemikalier som hovedsakelig blir brukt i industri og jordbruk. Disse giftstoffene blir transportert til områder med permafrost via luft- og havstrømmer fra sørlige breddegrader, men det finnes også noen lokale kilder. Når giftstoffene ankommer Arktis avhenger deres videre skjebne av deres fysiske og kjemiske egenskaper.

Snø spiller en viktig rolle i deponeringen av HOC giftstoffene. I snø kan HOC gå over til gassform igjen, bli kjemisk nedbrutt samt at lagringsevnen til snø svekkes etter hvert som den eldes, noe som fører til remobilisering av giftstoffene. Derfor er ikke konsentrasjonene av HOC i snø konstante gjennom sesongen, og mengden med giftstoffer i smeltevann er som oftest mindre enn den opprinnelige mengden i snøen. Isbreer formes og vokser som et resultat av akkumulering av snø, og dermed gjelder prosessene beskrevet ovenfor også for isbreer. HOC-nivåer er generelt lave i permafrostjord da grunnen som regel fortsatt er frosset når snøen smelter, og giftstoffene blir transportert i smeltevann til vassdrag. Et unntak er jord i og nærrområder med lokale kilder hvor det kan være betydelige konsentrasjoner av HOC i grunnmaterialet. På Svalbard er de høyeste nivåene av PCB i jord funnet i de russiske bosettingene, hvor konsentrasjonen er cirka hundre ganger høyere enn bakgrunnsverdier.

Klimaendringer kan føre til økt deponering av HOC på landområder med permafrost fordi det gir økt regn over landområder. Dette skjer muligens på Svalbard hvor vintertemperaturer er høyere og nedbør kommer mer som regn enn snø enn tidligere. På bakgrunn av den globalt økende permafrosttemperaturen og utvidelsen av dybden på det aktive laget, noe som også skjer på Svalbard, er faren at giftstoffer i permafrost skal forflytte seg fra jord (og vann og is) til luft. Stigende temperaturer kan også lede til økt mobilisering av HOC fra forurenset jord. Smelting av isbreer kan dessuten føre til økt utslipp av HOC til luft, sjø og jord.

Arctic permafrost areas

Permafrost is ground (soil, rock and/or sediment) that is 0 °C or colder for at least two consecutive years (French, 2013, AMAP, 2011). The distribution of permafrost is widespread in the northern hemisphere (12.8-17.8% of exposed land is underlain with permafrost (Zhang et al., 2000)), with the majority found in the Arctic (AMAP, 2011). The thickness of permafrost varies considerably, from just centimeters to 1500 m in the high Arctic (AMAP, 2011). There are four categories of permafrost: continuous (90-100% of area has permafrost), discontinuous (50-90%), sporadic (10-50%) and isolated (0-10%) (French, 2013).

Permafrost in Svalbard is extensive and continuous with a thickness of 100 m in the valleys and 400-500 m in the mountainous areas (Christiansen et al., 2010). A little less than 40% of Svalbard is underlain with permafrost as the ground beneath glaciers is usually above 0 °C (Humlum et al., 2003). On mainland Norway the permafrost is restricted to alpine areas and to northern Norway where the lower limit for permafrost is at 800-900 meters above sea level (m.a.s.l.) in the coastal areas of Troms, 400-500 m.a.s.l. in Finnmark and 600-700 m.a.s.l. inland (Isaksen et al., 2007b, Christiansen et al., 2010).

Above the permafrost is the active layer which thaws and freezes during summer and winter, respectively. The active layer is not part of the permafrost, as it reaches temperatures above 0 °C annually. Similar to permafrost, the thickness of the active layer varies regionally and seasonally depending on factors like snow cover and air temperature etc., from 50 cm to up to 10 m depending on the ground material (French, 2013).

With increasing air temperatures permafrost is thawing and permafrost temperatures have generally increased by 0.5-2 °C since the late 1970s (AMAP, 2011). There are large differences between areas due to variation in for example ocean currents, snow and rain precipitation, topography, vegetation and ground material (rocks and soils have different thermal conductivities and albedos (French, 2013)). In Scandinavia the air temperature was around 1 °C warmer during the International Polar Year than in the preceding decade (Christiansen et al., 2010, Romanovsky et al., 2010). Two studies have found indications that permafrost in Svalbard and northern Scandinavia is warming (Isaksen et al., 2007a, Isaksen et al., 2007b). Monitoring data from Svalbard show that temperature in the upper part of the permafrost has risen on average 0.7 °C per decade during 1998-2014 with an accelerating speed during the past decade (www.mosj.no). In addition, the active layer has become 25–30 cm thicker since 1998 (www.mosj.no).

The consequences of thawing permafrost include release of greenhouse gases, erosion, threat to infrastructure, release of viable ancient microbes, changes in ecosystems and release of contaminants (Isaksen et al., 2007a, Lemke et al., 2007, AMAP, 2011). With the increase in air temperature due to climate change, continuous permafrost will thaw and reduce in thickness, the active layer thickness increase, and discontinuous permafrost may disappear altogether (French, 2013).



Figure 1. Permafrost distribution in the Arctic. Map by Philippe Rekacewicz, UNEP/GRID-Arendal. http://www.grida.no/graphicslib/detail/permafrost-distribution-in-the-arctic_3823

Production and regulation of HOCs

Polychlorinated biphenyls (PCBs) were used as industrial chemicals from the 1940s until the early 1980s. The global production of PCBs has been estimated at approximately 1.3 million tons from 1930-1992 (de Wit et al., 2004). PCBs consist of 209 separate components with 1-10 chlorine atoms in the molecule. PCBs have been used in many products, such as electrical equipment, hydraulic systems, paint and insulation. Regulation of PCB started at the end of the 1970s and an international ban by the Stockholm Convention entered into force in 2004 (Stockholm Convention, 2001).

Organochlorinated pesticides (OCPs) are a group comprising many hundreds of compounds formerly used in agriculture to combat pests or weeds. As practically all these substances have been found to be harmful to the environment, stringent restrictions on their use and storage have been introduced. A number of chlorinated pesticides (aldrin, chlordane, DDT, dieldrin, endrin, heptachlorine, hexachlorobenzene [HCB], mirex and toxaphene) were banned in 2004 by the Stockholm Convention (Stockholm Convention, 2001). Several more pesticides were listed under the Convention in 2009 (chlordecone, alpha- and beta-hexachlorocyclohexane [HCH], lindane and pentachlorobenzene) (Stockholm Convention, 2009). The technical mixture of HCH was a common insecticide from the 1940s, and was used as a replacement for DDT in the 1960s and 1970s. The use of lindane, another HCH mixture, was permitted in Norway until 1992. In the year 2000, the use of technical HCH and lindane was forbidden in China and France, the principal nations using these mixtures at the end of the 1990s. Chlordanes are a group of substances formerly used to combat insects, for the most part in subtropical regions of the USA, Central America and Asia; they have been very little used in Europe. It is therefore assumed that atmospheric long-range transport of chlordanes largely comes from America and Asia. DDT was banned as an insecticide in the early 1970s. Due to the need to effectively combat a rise in malaria in southerly regions, DDT was again permitted in tropical countries, but subject to considerable restrictions on its use and discharge (UNEP/AMAP Expert Group, 2011). There are therefore still fresh sources of DDT, especially in connection with large-scale combatment of the malaria mosquito in Asia and Africa. HCB is a compound formerly used as a fungicide, but it now mostly occurs as an industrial by-product from the chemical industry. Its presence in the environment is therefore mainly due to industrial emissions (AMAP, 2014).

Brominated flame retardants (BFRs) have been in use since the 1970s to make products less inflammable. They have been used in building materials, electronic articles, furniture, motor vehicles plastics and textiles. The most commonly used compounds have been polybrominated biphenyls (PBB), polybrominated diphenyl ethers (PBDE), hexabromocyclododecane (HBCDD) and tetrabromobisphenol A (TBBPA). These BFRs are considered to have physical properties resembling the chlorinated organic pollutants (for example PCB and DDT). PBDEs including penta- and octa-brominated diphenyl ethers (BDEs) were added under the Stockholm Convention in 2009 (Stockholm Convention, 2009) and amendment listing of hexabromocyclododecane (HBCDD) has entered into force (Stockholm Convention, 2013). A directive on the restriction of penta- and octaBDE was adopted in 2003 by the European Union (European Parliament and the Council, 2003) whereas the production of these compounds was ceased some years later in North America (www.epa.gov) (Canadian Environmental Protection Act, 2008).

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) have been used since the 1950s for various industrial purposes such as fire fighting foam, textiles, photo imaging, hydraulic fluids, electric and electronic parts. Perfluorooctane sulfonyl fluoride (PFOSF) based compounds, including perfluorooctane sulfonate (PFOS), were gradually phased out beginning in the year 2000 by 3M, which was the major producer (3M, 2000). Eight years later PFOSF-based compounds were included in annex B of the Stockholm Convention, which means that the production and use is restricted but not completely banned (Stockholm Convention, 2009). However, manufacturing of PFOS and related substances started at high quantities in China in the early 2000s (Wang et al., 2014, Xie et al., 2013). The production

and use of perfluorooctanoate (PFOA), which belongs to perfluoroalkyl carboxylates (PFCAs), and similar compounds with a longer carbon chain have gradually been phased out in the United States following agreements with major manufacturers (EPA, 2006). PFCAs emissions by Western Europe and Japan have also decreased since 2000 (Wang et al., 2014). PFCAs with eight or more carbons are currently considered for the candidate list of substances of very high concern for authorization by the European Chemicals Agency (<http://echa.europa.eu/en/candidate-list-table>). In addition, PFOA and its precursors are proposed for listing under the Stockholm Convention (Stockholm Convention, 2015). However, production of PFCAs and their precursors has dramatically increased since 2002 in China and other developing Asian economies (Wang et al., 2014).

Sources of HOCs in Arctic permafrost areas

Most permafrost areas are far away from industrial areas. The presence of contaminants in pristine Arctic areas is thus mostly due to long-range transport although local sources may also be present. HOCs may be transported to the Arctic by air and ocean currents from southern industrialized areas. PCBs, most chlorinated pesticides and brominated compounds are mainly subject to long-range transport by air currents, whereas the main transport routes for beta-hexachlorocyclohexane (β -HCH) are ocean currents (Frank and Dugani, 2003, Wania and Mackay, 1993, Li and Macdonald, 2005, Macdonald et al., 2005b). The deposition of atmospherically transported compounds to the surface is facilitated by cold-condensation in Arctic areas (Ma, 2010, Wania and Mackay, 1993). This means that as the temperature falls, chemicals in the atmosphere "condense" onto soil, water, aerosols, snow and ice. Volatile precursors of PFAS may undergo long-range atmospheric transport and degrade to PFASs and PFCAs in the atmosphere and snow (Ellis et al., 2004, Martin et al., 2006, D'Eon et al., 2006, Taniyasu et al., 2013), whereas PFASs and PFCAs may reach the Arctic via ocean currents after being directly released to the aquatic environment (Zhao et al., 2012, Wania, 2007). From the atmosphere to the surface PFAS are transported mainly by wet deposition like rain and snow (Taniyasu et al., 2013, Stemmler and Lammel, 2010).

Local pollution may also be a source for HOCs in Arctic permafrost areas. Examples of point sources are remote military stations at Saglek in Labrador, and Thule in Greenland (Kuzyk et al., 2005, Glahder et al., 2003). Poor handling and disposal of transformer oil waste during the 1950s-1970s resulted in elevated PCB concentrations in both terrestrial and marine ecosystems in the proximity of Saglek. Elevated concentrations of PCBs in terrestrial sites were highest within a few kilometers range but decreased exponentially with distance from the contaminated sites (Pier et al., 2003, Kuzyk et al., 2005). In Svalbard, elevated PCB levels have been observed in the proximity of the Russian settlements Barentsburg and Pyramiden (Jartun et al., 2009). Furthermore, local airports may also act as PFAS sources in remote Arctic areas (Lescord et al., 2015). Monitoring data from Svalbard show decreasing air concentrations of PCBs, HCH, chlordanes and DDT (www.mosj.no). In contrast, HCB concentrations have increased during the last decade (www.mosj.no).

Fate of HOCs in Arctic permafrost areas

Once the HOCs reach the Arctic, their fate depends on their physico-chemical properties as well as the surrounding environmental conditions (Mackay and Wania, 1995, Wania and Mackay, 1995). Chemicals transported through the atmosphere can be deposited to the ground, either by dry or wet deposition (Grannas et al., 2013) and they are consequently found in snow, glacier ice, meltwater and soil.

Snow

Snow is an efficient scavenger of atmospheric contaminants. As snowfall typically constitutes more than half of annual precipitation in the Arctic, snow is an important factor in the fate of the HOCs (Franz and Eisenreich, 1998, Grannas et al., 2013).

Thus, snowfall leads to wet deposition of HOCs and accumulation of HOCs in snowpack. Concentrations of HOCs detected in snow across the Arctic vary from pg/L to ng/L (Macdonald et al., 2000, Melnikov et al., 2003, Herbert et al., 2005, Young et al., 2007). HOC concentrations in snow do not necessarily reflect initial deposition, as the concentrations may be influenced by sampling period and method, wind speed, temperature and age of snow among other factors (Gregor et al., 1996, Wania et al., 1998). This is illustrated in Figure 2.

HOCs deposited in the snowpack can undergo several processes through which they are re-mobilized. As the snowpack ages, the surface area of ice crystals decreases resulting in a reduction in storage capacity. Consequently, contaminants desorb and enter the interstitial air with subsequent release to the atmosphere (Cabanés et al., 2003). Herbert et al. (2005) studied POP concentrations in snow from Tromsø and reported that PCB concentrations decreased by ~75% within the first 96 hours. Similarly, α - and γ -HCH were reduced from 380 to 30 pg/L and from 2000 to 270 pg/L, respectively, in the first 96 hours of the sampling period. Wind action may further enhance the snow—atmosphere exchange by ventilating the interstitial air within the snowpack where HOCs may be present as gases (Albert et al., 2002, Halsall, 2004).

Highly volatile compounds are quicker re-volatilized into the atmosphere compared to the less volatile ones, and some compounds may also be chemically degraded in the snowpack (Grannas et al., 2013). HOC revolatilization from snowpack is also related to rainfall. A rainfall event may form a frozen layer on top of the snowpack. As a result of this, contaminants are effectively sealed in, and a larger proportion of contaminants than usual are released in the meltwater (Grannas et al., 2013). This is becoming more common in the Arctic as the climate is warming (Hansen et al., 2014). Nevertheless, it is primarily the most photochemically stable and least volatile HOCs that dominate the contaminant load in the snowpack at the time of melting.

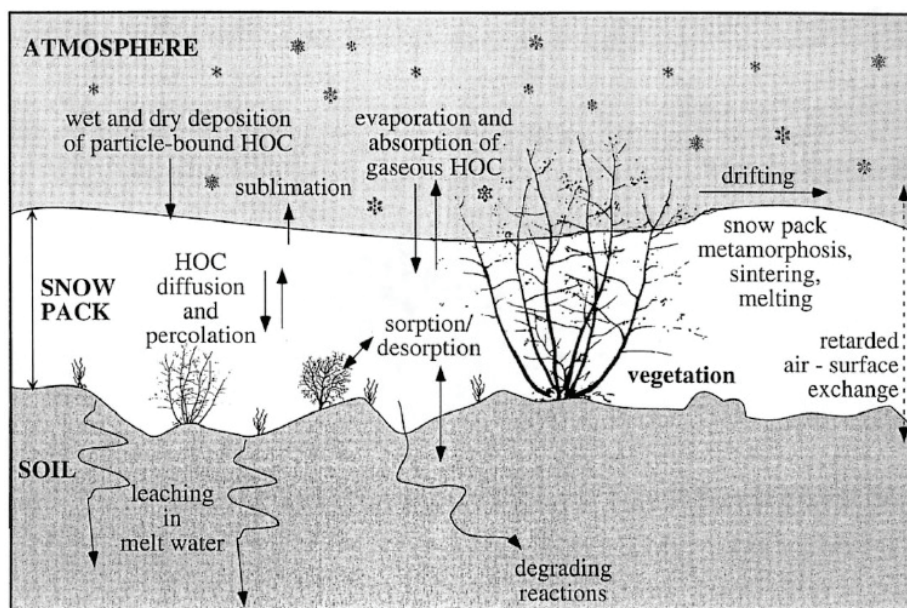


Figure 2. Fate of halogenated organic contaminants (HOCs) in snow, from Wania et al. (1998; reprinted with permission).

Glacier ice

Glaciers are formed and grow when the accumulation of snow in winter is greater than the loss of snow in summer. The remaining snow is transformed into firn and is gradually compressed and changed into ice by the pressure of overlying snow (Hambrey and Alean, 2004). Ice cores can be extracted from the glacier and dated, providing an opportunity to study past atmospheric deposition (Donald et al., 1999, Garmash et al., 2013, Kwok et al., 2013). However, the accuracy of the ice core-method is arguable, as contaminants may be re-volatilized and degraded in the snow (Grannas et al., 2013).

As glaciers are mainly composed of snow, the mechanisms and pathways for contaminant deposition discussed above are also relevant here. Contaminants are deposited onto the glacier through wet and dry deposition. HOCs in the snow which does not melt will then be available for integration into the glacier.

A study by Garmash et al. (2013) reported PCB fluxes from a 37 m ice core representing the time period 1957 - 2009 from Lomonosovfonna, Svalbard. While the \sum PCB flux varied over time, the largest fluxes occurred during 1957-1966, 1974-1983, 1998-2009 and 2009-2010 with $\sim 19 \text{ pg cm}^{-2} \text{ y}^{-1}$. Donald et al. (1999) analysed samples from a glacier in the Canadian Rocky Mountains for organochlorine pesticides. They found a maximum concentration of 2.57 ng/L for \sum DDT in 1983 and 0.054 ng/L for dieldrin in 1989. \sum Endosulfan and \sum chlordanes both peaked in 1989 with 0.41 ng/L and 0.066 ng/L, respectively. The greatest concentrations detected for lindane and α -HCH was 0.12 ng/L and 1.82 ng/L, both from 1961. Hermanson et al. (2010) examined brominated flame retardants (BFRs) in a 34 m ice core from the glacier Holtedahlfonna in Svalbard. Of the BFRs most commonly detected, HBCD and BDE-209 were found in the greatest concentrations. The inputs of HBCD and BDE-209 were largest in the segment of ice corresponding to atmospheric deposition between 1995 and 2005 with $910 \text{ pg cm}^{-2} \text{ y}^{-1}$ and $320 \text{ pg cm}^{-2} \text{ y}^{-1}$, respectively. Concentrations of PFASs were investigated in the Longyearbreen glacier by Kwok et al. (2013). The dominant compounds detected were PFBA, PFOA and PFNA.

Meltwater

As glaciers are receding worldwide (Dowdeswell et al., 1997, Yao et al., 2007, Ye et al., 2007, Farinotti et al., 2009, Jacobs et al., 2011), contaminants stored in glaciers may be released with meltwater. For example, Bogdal et al. (2009) observed the release of POPs from melting alpine glaciers into Lake Oberaar, Switzerland, by analysing a dated sediment core. Similarly, Bizzotto et al. (2009) reported higher concentrations of POPs in a glacier-fed stream than in a non-glacial stream from the same valley in the Italian Alps during the period of greatest glacial melt. Furthermore, in a study from the Canadian Rocky Mountains, 50-97% of POP inputs to a subalpine lake were from melting glaciers (Blais et al., 2001).

Glacial melt in the Arctic has likely little significance on the release of contaminants because Arctic glacial melt is mainly from zones where pre-industrial ice is emerging (Macdonald et al., 2005a). The meltwater would then be relatively clean and act to dilute contaminants released. The authors note, however, that melting glaciers may release significant concentrations of DDT in some Arctic locations.

The HOCs remaining within the snowpack at the onset of spring will be transported by the meltwater to receiving watersheds. As the ground is frozen, bottom-melting is negligible and the main mechanism by which the snowpack melts is surface-melting (Ling and Zhang, 2004, Meyer and Wania, 2008). With surface-melting, water is able to trickle down the snowpack and incorporate contaminants along the way resulting in a more contaminated meltwater than would be expected from bottom-melting meltwater (Meyer and Wania, 2008). As the snowpack is underlain by permafrost and the frozen ground is impermeable to water, the input of contaminated meltwater to the soil is relatively minor (Meyer and Wania, 2008).



Figure 3. Permafrost landscape at Kapp Linné, Svalbard. Photo: Geir Wing Gabrielsen, Norwegian Polar Institute.

Soil

In and around human settlements there are often elevated concentrations of contaminants in the soil due to human activity (Table 1). Waste handling and disposal are frequently considerable local sources of HOCs (Grannas et al., 2013). Indeed Danon-Schaffer et al. (2008) found very high levels of PBDEs in samples from landfill soils in the Canadian Arctic. With the warming ground, there is concern that waste contained within the permafrost may be able to undergo transport away from the landfill/dumpsite and affect the surrounding environment. Bright et al. (1995) investigated the Distant Early Warning (DEW) Line, which was a series of radar stations in northern Canada where the use of PCBs is well known. Soil samples were taken near the station and reference samples were taken from sites deemed to be pristine. PCB levels up to 1 000 000 ng/g were detected in the most heavily polluted areas. Quantifiable concentrations of PCB were also detected in the background samples, with two background sites having a mean PCB concentration of 14 and 28 ng/g, respectively, suggesting that PCBs go through short-range transport. Pier et al. (2003) also reported short-range transport of PCB in Saglek Bay, as PCB contamination spread from point-sources in soil up to 50 km.

In Svalbard, the background level of PCBs in soil is 1-5 ng/g (Jartun et al., 2010). Jartun et al. (2009) reported relatively high PCB concentrations in Russian coal mining settlements in Svalbard. Median PCB concentrations were 172 and 268 ng/g in soils from Pyramiden (an abandoned settlement), and Barentsburg (an inhabited settlement), respectively. In contrast, Longyearbyen had a median concentration of < 4 ng/g PCB in soil. Paint and electrical equipment were identified as the likely sources of pollution in the Russian settlements.

Zhu et al. (2015) investigated levels of PCBs and PBDEs in various media around Kongsfjorden, Svalbard. The mean concentrations in soil were 1.38 ng/g dw for PCBs and 0.0655 ng/g dw for PBDEs. The highest soil concentrations of both PCBs and PBDEs were found approximately 2 km southeast of the settlement of Ny-Ålesund. Similarly, Na et al. (2015) examined the concentrations of the flame retardants dechloranes in soil around Kongsfjorden. The mean concentration of \sum Dechlorane was 0.342 ng/g dw. As reported by Zhu et al. (2015), the highest soil concentration of dechloranes was southeast of Ny-Ålesund. Neither of the studies discuss why this area appears to be more contaminated.

A research station was established at Kinnvika, Svalbard during 1957-1958 and subsequently abandoned in 1958, with little use since. Evenset and Christensen examined soil concentrations of PCBs at Kinnvika, Svalbard, and presented their results in two separate reports. The minimum - maximum concentrations of \sum_7 PCB reported were <LOD - 32 ng/g dw and <LOD - 1.33 ng/g dw, respectively (Evenset and Christensen, 2011, Evenset and Christensen, 2012).

Bjørnøya is situated approximately halfway between mainland Norway and Spitsbergen and is part of the Svalbard archipelago. Similar to other sites in Svalbard, the island has a history of resource exploitation with past hunting activities, whaling and coal mining. Today there is some research activity and a manned meteorological station. There is not a lot of literature available for Bjørnøya, but Meijer et al. (2002) found relatively low levels (0.051, 0.055 and 0.057 ng/g dw) of \sum PCB at three locations. In contrast, Jartun et al. (2010) report a mean concentration of 26 ng/g \sum_{21} PCB based on nine soil samples from Bjørnøya. The large difference in PCB concentrations between the two studies is likely due to the fact that Meijer et al. (2002) intentionally sampled soil from areas away from human activities.

Table 1. HOC concentrations (ng/g) in soil samples from different localities in the Arctic.

HOC	Conc	Place	Year	Reference	Note
Σ_9 PBDE	180.7	Iqaluit, Canada	2004 - 2006	(Danon-Schaffer et al., 2008)	Landfill soil
Σ_9 PBDE	154.6	Cambridge Bay, Canada	2004 - 2006	(Danon-Schaffer et al., 2008)	Landfill soil
Σ_9 PBDE	9.10	Yellowknife, Canada	2004 - 2006	(Danon-Schaffer et al., 2008)	Landfill soil
Σ_{13} PBDE	0.0655	Kongsfjorden, Svalbard	2011 - 2012	(Zhu et al., 2015)	
Σ_3 PBDE	0.16 - 0.23	Russian Arctic	2000 - 2001	(de Wit et al., 2006)	
Σ_5 Dechlorane	0.342	Kongsfjorden, Svalbard	2012	(Na et al., 2015)	
Σ_7 PCB	1 - 5	Svalbard	2007 - 2009	(Jartun et al., 2010)	Background
Σ_{25} PCB	1.38	Kongsfjorden, Svalbard	2011 - 2012	(Zhu et al., 2015)	
Σ_7 PCB	26	Bjørnøya, Svalbard	2007 - 2009	(Jartun et al., 2010)	
Σ_{29} PCB	0.051	Bjørnøya, Svalbard	1998	(Meijer et al., 2002)	
Σ_{29} PCB	0.055	Bjørnøya, Svalbard	1998	(Meijer et al., 2002)	
Σ_{29} PCB	0.057	Bjørnøya, Svalbard	1998	(Meijer et al., 2002)	
Σ_7 PCB	32	Kinnvika, Svalbard	2011	(Evenset and Christensen, 2011)	
Σ_{64} PCB	1.33	Kinnvika, Svalbard	2011	(Evenset and Christensen, 2012)	
Σ_7 PCB	268	Barentsburg, Svalbard	2009	(Jartun et al., 2009)	
Σ_7 PCB	172	Pyramiden, Svalbard	2009	(Jartun et al., 2009)	
Σ_7 PCB	< 4.0	Longyearbyen, Svalbard	2009	(Jartun et al., 2009)	
Σ PCB	1 000 000	DEW Station, Canada	1989 - 1992	(Bright et al., 1995)	Polluted site
Σ PCB	14	Canadian Arctic	1989 - 1992	(Bright et al., 1995)	Background
Σ PCB	250 000	Saglek Bay, Canada	1997 - 1999	(Pier et al., 2003)	Polluted site

Degradation of HOCs in Arctic permafrost areas

Photodegradation is a major route by which contaminants are degraded, both in the atmosphere and once they are deposited into snow/ice (Klán and Holoubek, 2002, Grannas et al., 2007, McNeill et al., 2012). The chemical reactions in snow/ice can either be photochemically or thermally induced (Grannas et al., 2013).

Higher chlorinated PCBs may be subject to microbial dechlorination where lesser chlorinated congeners may be oxidized to OH-PCBs in the atmosphere or by microbes (Tehrani and Van Aken, 2014). However, OH-PCB formation by microbes is generally low (Tehrani and Van Aken, 2014). Fluorotelomer alcohols (FTOHs) and perfluoroalkyl sulfonamides (PFOSA) may degrade to PFASs and PFCAs in atmosphere and snow (Ellis et al., 2004, Martin et al., 2006, D'Eon et al., 2006, Taniyasu et al., 2013) and biotransformation of PFAS also occurs by soil microbes (Butt et al., 2014). It is expected that microbial degradation in Arctic soils is relatively low as microbial enzyme activities in Arctic soil is somewhat lower compared to other ecosystems (Wallenstein et al., 2009). Furthermore, enzymes activities are highest

before the period of snow melt and subsequent release of contaminants from the snow pack (Wallenstein et al., 2009).

Impact of climate change on HOCs in Arctic permafrost areas

Deposition of HOCs on permafrost land areas may augment with increased rain over land (Meyer and Wania, 2007). The chemicals that are affected by rain rate include compounds that have high affinity towards soil, such as lindane, aldrin, highly chlorinated PCBs, PBDEs and some currently used pesticides (Meyer and Wania, 2007). However, under warming temperatures, HOCs will be more prone to move from soil and other depository compartments (water and ice) to the air after deposition (Ma and Cao, 2010). Already, a wide range of HOCs have been remobilized from water, soil and ice into the Arctic atmosphere over the past two decades as a result of climate change (Ma et al., 2011).

Implications for Svalbard

Svalbard is already affected by the ramifications of a changing climate. The winters have become warmer with more precipitation falling as rain (Hansen et al., 2014), the temperature of the permafrost has increased (Isaksen et al., 2007a, Isaksen et al., 2007b) and glaciers are receding (Dowdeswell et al., 1997, Hagen et al., 2003, Błaszczuk et al., 2009).

As discussed in the section above, this will likely lead to a greater deposition of certain HOCs into snow and soil, although their re-volatilization will also be enhanced. There may be an increase in HOC concentrations in meltwater due to rain-on-snow events becoming more frequent, sealing the contaminants in the snowpack (Grannas et al., 2013, Hansen et al., 2014). Some of the HOCs present in the snowpack at the time of melting will move into the thawing active layer, while most will be transported in the meltwater to receiving drainage basins.

With the warming permafrost, the active layer will increase in depth and result in an enhanced mobility for previously contained contaminants. However, contaminant concentrations are generally low in soil except at sites close to local sources. Furthermore, polluted sites (e.g. areas within Pyramiden and Barentsburg) that benefit from the fact that the impermeability of permafrost immobilize contaminants, may experience a change in the spatial profile of the pollution. This, in turn, makes formerly inaccessible HOCs potentially bioavailable again to flora and fauna.

The shrinking glaciers will release stored HOCs, but as previously discussed, the extent of this depends on which parts of the glaciers are melting and the degree of contamination of those (Macdonald et al., 2005a). Nevertheless, Kwok et al. (2013) report higher concentrations of PFASs in glacial meltwater compared to glacier ice samples. However, they also note that the level of PFASs increase closer to the settlement of Longyearbyen and thus local sources of pollution are likely of greater significance than glacial meltwater for current-use HOCs.

Conclusion

Halogenated organic contaminants are present in Arctic permafrost areas primarily due to long-range transport, either via air or ocean currents. Once they reach the Arctic, they can be deposited on soil, snow and glaciers. High contaminant concentrations have been reported in the proximity of a few local pollution sources, such as military stations. HOCs at the permafrost surface can be remobilized and enter the atmosphere or be degraded. With the changing climate, more HOCs may be deposited with increasing rainfall events. Also, glacial meltwater may release contaminants hitherto stored in ice and the warming permafrost will increase the mobility of HOCs within the soil.

References

- 3M 2000. Re: phase-out plan for POSF-based products (226–0600). *US EPA Adm Rec.*, 226, 1-11.
- ALBERT, M. R., GRANNAS, A. M., BOTTENHEIM, J., SHEPSON, P. B. & PERRON, F. E. 2002. Processes and properties of snow–air transfer in the high Arctic with application to interstitial ozone at Alert, Canada. *Atmospheric Environment*, 36, 2779-2787.
- AMAP 2011. *Snow, Water, Ice and Permafrost in the Arctic (SWIPA): Climate Change and the Cryosphere*, Arctic Monitoring and Assessment Programme (AMAP).
- AMAP 2014. Trends in Stockholm Convention Persistent Organic Pollutants (POPs) in Arctic Air, Human Media and Biota. *AMAP Technical Report*. AMAP: Arctic Monitoring and Assessment Programme.
- BIZZOTTO, E., VILLA, S., VAJ, C. & VIGHI, M. 2009. Comparison of glacial and non-glacial-fed streams to evaluate the loading of persistent organic pollutants through seasonal snow/ice melt. *Chemosphere*, 74, 924-930.
- BLAIS, J. M., SCHINDLER, D. W., MUIR, D. C., SHARP, M., DONALD, D., LAFRENIERE, M., BRAEKEVELT, E. & STRACHAN, W. M. 2001. Melting glaciers: a major source of persistent organochlorines to subalpine Bow Lake in Banff National Park, Canada. *AMBIO: A Journal of the Human Environment*, 30, 410-415.
- BŁASZCZYK, M., JANIA, J. A. & HAGEN, J. O. 2009. Tidewater glaciers of Svalbard: Recent changes and estimates of calving fluxes. *Polish Polar Research*, 30, 85-142.
- BOGDAL, C., SCHMID, P., ZENNEGG, M., ANSELMETTI, F. S., SCHERINGER, M. & HUNGERBÜHLER, K. 2009. Blast from the past: melting glaciers as a relevant source for persistent organic pollutants. *Environmental Science & Technology*, 43, 8173-8177.
- BRIGHT, D. A., DUSHENKO, W. T., GRUNDY, S. L. & REIMER, K. J. 1995. Evidence for short-range transport of polychlorinated biphenyls in the Canadian Arctic using congener signatures of PCBs in soils. *Science of the Total Environment*, 160, 251-263.
- BUTT, C. M., MUIR, D. C. G. & MABURY, S. A. 2014. Biotransformation pathways of fluorotelomer-based polyfluoroalkyl substances: a review. *Environmental Toxicology and Chemistry*, 33, 243-267.
- CABANES, A., LEGAGNEUX, L. & DOMINÉ, F. 2003. Rate of evolution of the specific surface area of surface snow layers. *Environmental Science & Technology*, 37, 661-666.
- CANADIAN ENVIRONMENTAL PROTECTION ACT. 2008. Polybrominated Diphenyl Ethers Regulations SOR/2008-218.
- CHRISTIANSEN, H. H., ETZELMÜLLER, B., ISAKSEN, K., JULIUSSEN, H., FARBROT, H., HUMLUM, O., JOHANSSON, M., INGEMAN-NIELSEN, T., KRISTENSEN, L. & HJORT, J. 2010. The thermal state of permafrost in the Nordic area during the International Polar Year 2007–2009. *Permafrost and Periglacial Processes*, 21, 156-181.
- D'EON, J. C., HURLEY, M. D., WALLINGTON, T. J. & MABURY, S. A. 2006. Atmospheric chemistry of N-methyl perfluorobutane sulfonamidoethanol, C₄F₉SO₂N(CH₃)CH₂CH₂OH: Kinetics and mechanism of reaction with OH. *Environmental Science & Technology*, 40, 1862-1868.

- DANON-SCHAFFER, M., GRACE, J. & IKONOMOU, M. 2008. PBDEs in waste disposal sites from Northern Canada. *Organohalogen Compounds*, 70, 365-368.
- DE WIT, C., FISK, A. T., HOBBS, K. E., MUIR, D. C. G., GABRIELSEN, G. W., KALLENBORG, R., KRAHN, M. M., NORSTROM, R. J. & SKAARE, J. U. 2004. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic. Oslo, Norway: Arctic Monitoring and Assessment Programme (AMAP).
- DE WIT, C. A., ALAEE, M. & MUIR, D. C. 2006. Levels and trends of brominated flame retardants in the Arctic. *Chemosphere*, 64, 209-233.
- DONALD, D. B., SYRGIANNIS, J., CROSLY, R. W., HOLDSWORTH, G., MUIR, D. C., ROSENBERG, B., SOLE, A. & SCHINDLER, D. W. 1999. Delayed deposition of organochlorine pesticides at a temperate glacier. *Environmental Science & Technology*, 33, 1794-1798.
- DOWDESWELL, J. A., HAGEN, J. O., BJÖRNSSON, H., GLAZOVSKY, A. F., HARRISON, W. D., HOLMLUND, P., JANIA, J., KOERNER, R. M., LEFAUCONNIER, B. & OMMANNEY, C. S. L. 1997. The mass balance of circum-Arctic glaciers and recent climate change. *Quaternary research*, 48, 1-14.
- ELLIS, D. A., MARTIN, J. W., DE SILVA, A. O., MABURY, S. A., HURLEY, M. D., ANDERSEN, M. P. S. & WALLINGTON, T. J. 2004. Degradation of fluorotelomer alcohols: A likely atmospheric source of perfluorinated carboxylic acids. *Environmental Science & Technology*, 38, 3316-3321.
- EPA 2006. Long-Chain Perfluorinated Chemicals (PFCs) Action Plan.
http://www.epa.gov/sites/production/files/2016-01/documents/pfcs_action_plan1230_09.pdf.
- EUROPEAN PARLIAMENT AND THE COUNCIL. 2003. Directive 2002/95/EC on the restriction of the use of certain hazardous substances in electrical and electronic equipment OJ L 37.
- EVENSET, A. & CHRISTENSEN, G. N. 2011. Undersøkelse av forurensningstilstand i Kinnvika, Nordaustlandet, Svalbard, 2011. Akvaplan-niva AS Rapport 5524-1.
- EVENSET, A. & CHRISTENSEN, G. N. 2012. Miljøgifter i jord, marine sedimenter og fisk fra Kinnvika, Nordaustlandet, Svalbard, 2011. Akvaplan-niva AS Rapport 5203 - 1.
- FARINOTTI, D., HUSS, M., BAUDER, A. & FUNK, M. 2009. An estimate of the glacier ice volume in the Swiss Alps. *Global and Planetary Change*, 68, 225-231.
- FRANK, W. & DUGANI, C. B. 2003. Assessing the long-range transport potential of polybrominated diphenyl ethers: a comparison of four multimedia models. *Environmental Toxicology and Chemistry*, 22, 1252-1261.
- FRANZ, T. P. & EISENREICH, S. J. 1998. Snow scavenging of polychlorinated biphenyls and polycyclic aromatic hydrocarbons in Minnesota. *Environmental Science & Technology*, 32, 1771-1778.
- FRENCH, H. M. 2013. *The periglacial environment*, John Wiley & Sons.
- GARMASH, O., HERMANSON, M. H., ISAKSSON, E., SCHWIKOWSKI, M., DIVINE, D., TEIXEIRA, C. & MUIR, D. C. 2013. Deposition history of polychlorinated biphenyls to the Lomonosovfonna glacier, Svalbard: A 209 congener analysis. *Environmental Science & Technology*, 47, 12064-12072.
- GLAHDER, C. M., ASMUND, G., MAYER, P., LASSEN, P., STRAND, J. & RIGET, F. 2003. Marin recipientundersøkelse ved Thule Air Base 2002. *Faglig rapport fra DMU*. Danmarks Miljøundersøgelser.
- GRANNAS, A., BOGDAL, C., HAGEMAN, K., HALSALL, C., HARNER, T., HUNG, H., KALLENBORN, R., KLÁN, P., KLÁNOVÁ, J. & MACDONALD, R. 2013. The role of the global cryosphere in the fate of organic contaminants. *Atmospheric Chemistry and Physics*, 13, 3271-3305.
- GRANNAS, A., JONES, A. E., DIBB, J., AMMANN, M., ANASTASIO, C., BEINE, H., BERGIN, M., BOTTENHEIM, J., BOXE, C. & CARVER, G. 2007. An overview of snow photochemistry: evidence, mechanisms and impacts. *Atmospheric Chemistry and Physics*, 7, 4329-4373.
- GREGOR, D., TEIXEIRA, C. & ROWSELL, R. 1996. Deposition of atmospherically transported polychlorinated biphenyls in the Canadian Arctic. *Chemosphere*, 33, 227-244.
- HAGEN, J. O., MELVOLD, K., PINGLOT, F. & DOWDESWELL, J. A. 2003. On the net mass balance of the glaciers and ice caps in Svalbard, Norwegian Arctic. *Arctic, Antarctic, and Alpine Research*, 35, 264-270.

- HALSALL, C. J. 2004. Investigating the occurrence of persistent organic pollutants (POPs) in the arctic: their atmospheric behaviour and interaction with the seasonal snow pack. *Environmental Pollution*, 128, 163-175.
- HAMBREY, M. J. & ALEAN, J. 2004. *Glaciers*, Cambridge Univ Press.
- HANSEN, B. B., ISAKSEN, K., BENESTAD, R. E., KOHLER, J., PEDERSEN, Å. Ø., LOE, L. E., COULSON, S. J., LARSEN, J. O. & VARPE, Ø. 2014. Warmer and wetter winters: characteristics and implications of an extreme weather event in the High Arctic. *Environmental Research Letters*, 9, 114021.
- HERBERT, B., HALSALL, C., VILLA, S., JONES, K. & KALLENBORN, R. 2005. Rapid changes in PCB and OC pesticide concentrations in Arctic snow. *Environmental Science & Technology*, 39, 2998-3005.
- HERMANSON, M. H., ISAKSSON, E., FORSSTRÖM, S., TEIXEIRA, C., MUIR, D. C., POHJOLA, V. A. & VAN DE WAL, R. S. 2010. Deposition history of brominated flame retardant compounds in an ice core from Holtedahlfonna, Svalbard, Norway. *Environmental Science & Technology*, 44, 7405-7410.
- HUMLUM, O., INSTANES, A. & SOLLID, J. L. 2003. Permafrost in Svalbard: a review of research history, climatic background and engineering challenges. *Polar research*, 22, 191-215.
- ISAKSEN, K., BENESTAD, R., HARRIS, C. & SOLLID, J. 2007a. Recent extreme near-surface permafrost temperatures on Svalbard in relation to future climate scenarios. *Geophysical Research Letters*, 34.
- ISAKSEN, K., SOLLID, J. L., HOLMLUND, P. & HARRIS, C. 2007b. Recent warming of mountain permafrost in Svalbard and Scandinavia. *Journal of Geophysical Research: Earth Surface*, 112.
- JACOBS, S. S., JENKINS, A., GIULIVI, C. F. & DUTRIEUX, P. 2011. Stronger ocean circulation and increased melting under Pine Island Glacier ice shelf. *Nature Geoscience*, 4, 519-523.
- JARTUN, M., EGGEN, O. & OTTESEN, R. T. 2010. PCB fra lokale kilder på Svalbard - overflatejord og produkter 2007-2009. Norges Geologiske Undersøkelse. Rapport 2010.038
- 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. *Journal of Toxicology and Environmental Health, Part A*, 72, 284-294.
- KLÁN, P. & HOLOUBEK, I. 2002. Ice (photo) chemistry. Ice as a medium for long-term (photo) chemical transformations—environmental implications. *Chemosphere*, 46, 1201-1210.
- KUZYK, Z. A., STOW, J. P., BURGESS, N. M., SOLOMON, S. M. & REIMER, K. J. 2005. PCBs in sediments and the coastal food web near a local contaminant source in Saglek Bay, Labrador. *Science of the Total Environment*, 351–352, 264-284.
- KWOK, K. Y., YAMAZAKI, E., YAMASHITA, N., TANIYASU, S., MURPHY, M. B., HORII, Y., PETRICK, G., KALLERBORN, R., KANNAN, K. & MURANO, K. 2013. Transport of Perfluoroalkyl substances (PFAS) from an arctic glacier to downstream locations: implications for sources. *Science of the Total Environment*, 447, 46-55.
- LEMKE, P., REN, J., ALLEY, R. B., ALLISON, I., CARRASCO, J., FLATO, G., FUJII, Y., KASER, G., MOTE, P. & THOMAS, R. H. 2007. Observations: changes in snow, ice and frozen ground. Chapter 4 in S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor and H.L. Miller (eds.), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- LESCORD, G. L., KIDD, K. A., DE SILVA, A. O., WILLIAMSON, M., SPENCER, C., WANG, X. & MUIR, D. C. 2015. Perfluorinated and polyfluorinated compounds in lake food webs from the Canadian high arctic. *Environmental Science & Technology*, 49, 2694-2702.
- 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. *Science of the Total Environment*, 342, 87-106.
- LING, F. & ZHANG, T. 2004. A numerical model for surface energy balance and thermal regime of the active layer and permafrost containing unfrozen water. *Cold Regions Science and Technology*, 38, 1-15.

- MA, J. 2010. Atmospheric transport of persistent semi-volatile organic chemicals to the Arctic and cold condensation in the mid-troposphere – Part 1: 2-D modeling in mean atmosphere. *Atmospheric Chemistry and Physics*, 10, 7303-7314.
- MA, J. & CAO, Z. 2010. Quantifying the perturbations of persistent organic pollutants Induced by climate change. *Environmental Science & Technology*, 44, 8567-8573.
- MA, J., HUNG, H., TIAN, C. & KALLENBORN, R. 2011. Revolatilization of persistent organic pollutants in the Arctic induced by climate change. *Nature Climate Change*, 1, 255-260.
- MACDONALD, R., BARRIE, L., BIDLAMAN, T., DIAMOND, M., GREGOR, D., SEMKIN, R., STRACHAN, W., LI, Y., WANIA, F. & ALAEE, M. 2000. Contaminants in the Canadian Arctic: 5 years of progress in understanding sources, occurrence and pathways. *Science of the Total Environment*, 254, 93-234.
- MACDONALD, R., HARNER, T. & FYFE, J. 2005a. Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. *Science of the Total Environment*, 342, 5-86.
- MACDONALD, R. W., HARNER, T. & FYFE, J. 2005b. Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. *Science of the Total Environment*, 342, 5-86.
- MACKAY, D. & WANIA, F. 1995. Transport of contaminants to the Arctic: partitioning, processes and models. *Science of the Total Environment*, 160, 25-38.
- MARTIN, J. W., ELLIS, D. A., MABURY, S. A., HURLEY, M. D. & WALLINGTON, T. J. 2006. Atmospheric chemistry of perfluoroalkanesulfonamides: Kinetic and product studies of the OH radical and Cl atom initiated oxidation of N-ethyl perfluorobutanesulfonamide. *Environmental Science & Technology*, 40, 864-872.
- MCNEILL, V. F., GRANNAS, A. M., ABBATT, J. P., AMMANN, M., ARIYA, P., BARTELS-RAUSCH, T., DOMINE, F., DONALDSON, D. J., GUZMAN, M. I. & HEGER, D. 2012. Organics in environmental ices: sources, chemistry, and impacts. *Atmospheric Chemistry and Physics*, 12, 9653-9678.
- MEIJER, S., STEINNES, E., OCKENDEN, W. & JONES, K. C. 2002. Influence of environmental variables on the spatial distribution of PCBs in Norwegian and UK soils: implications for global cycling. *Environmental Science & Technology*, 36, 2146-2153.
- MELNIKOV, S., CARROLL, J., GORSHKOV, A., VLASOV, S. & DAHLE, S. 2003. Snow and ice concentrations of selected persistent pollutants in the Ob–Yenisey River watershed. *Science of the Total Environment*, 306, 27-37.
- MEYER, T. & WANIA, F. 2007. What environmental fate processes have the strongest influence on a completely persistent organic chemical's accumulation in the Arctic? *Atmospheric Environment*, 41, 2757-2767.
- MEYER, T. & WANIA, F. 2008. Organic contaminant amplification during snowmelt. *Water research*, 42, 1847-1865.
- NA, G., WEI, W., ZHOU, S., GAO, H., MA, X., QIU, L., GE, L., BAO, C. & YAO, Z. 2015. Distribution characteristics and indicator significance of Dechloranes in multi-matrices at Ny-Ålesund in the Arctic. *Journal of Environmental Sciences*, 28, 8-13.
- PIER, M. D., BETTS-PIPER, A. A., KNOWLTON, C. C., ZEEB, B. A. & REIMER, K. J. 2003. Redistribution of polychlorinated biphenyls from a local point source: terrestrial soil, freshwater sediment, and vascular plants as indicators of the halo effect. *Arctic, Antarctic, and Alpine Research*, 35, 349-360.
- ROMANOVSKY, V. E., SMITH, S. L. & CHRISTIANSEN, H. H. 2010. Permafrost thermal state in the polar Northern Hemisphere during the international polar year 2007–2009: a synthesis. *Permafrost and Periglacial Processes*, 21, 106-116.
- STEMMLER, I. & LAMMEL, G. 2010. Pathways of PFOA to the Arctic: variabilities and contributions of oceanic currents and atmospheric transport and chemistry sources. *Atmospheric Chemistry and Physics*, 10, 9965-9980.
- STOCKHOLM CONVENTION. 2001. UNEP/POPS/CONF/4. Final act of the conference of plenipotentiaries on the Stockholm Convention on persistent organic pollutants. Stockholm

- STOCKHOLM CONVENTION. 2009. C.N.524.2009.TREATIES-4 (Depositary Notification). Geneva
- STOCKHOLM CONVENTION. 2013. C.N.934.2013.TREATIES-XXVII.15 (Depositary Notification). Geneva
- STOCKHOLM CONVENTION. 2015. UNEP/POPS/POPRC.11/5: Proposal to list pentadecafluorooctanoic acid (CAS No: 335-67-1, PFOA, perfluorooctanoic acid), its salts and PFOA-related compounds in Annexes A, B and/or C to the Stockholm Convention on Persistent Organic Pollutants. Rome.
- TANIYASU, S., YAMASHITA, N., MOON, H. B., KWOK, K. Y., LAM, P. K. S., HORII, Y., PETRICK, G. & KANNAN, K. 2013. Does wet precipitation represent local and regional atmospheric transportation by perfluorinated alkyl substances? *Environment International*, 55, 25-32.
- TEHRANI, R. & VAN AKEN, B. 2014. Hydroxylated polychlorinated biphenyls in the environment: sources, fate, and toxicities. *Environmental Science and Pollution Research*, 21, 6334-6345.
- WALLENSTEIN, M. D., MCMAHON, S. K. & SCHIMMEL, J. P. 2009. Seasonal variation in enzyme activities and temperature sensitivities in Arctic tundra soils. *Global Change Biology*, 15, 1631-1639.
- WANG, Z., COUSINS, I. T., SCHERINGER, M., BUCK, R. C. & HUNGERBUEHLER, K. 2014. Global emission inventories for C-4-C-14 perfluoroalkyl carboxylic acid (PFCA) homologues from 1951 to 2030, Part I: production and emissions from quantifiable sources. *Environment International*, 70, 62-75.
- WANIA, F. 2007. A global mass balance analysis of the source of perfluorocarboxylic acids in the Arctic ocean. *Environmental Science & Technology*, 41, 4529-4535.
- WANIA, F., HOFF, J., JIA, C. & MACKAY, D. 1998. The effects of snow and ice on the environmental behaviour of hydrophobic organic chemicals. *Environmental Pollution*, 102, 25-41.
- WANIA, F. & MACKAY, D. 1993. Global fractionation and cold condensation of low volatility organochlorine compounds in polar-regions. *Ambio*, 22, 10-18.
- WANIA, F. & MACKAY, D. 1995. A global distribution model for persistent organic chemicals. *Science of the Total Environment*, 160, 211-232.
- XIE, S. W., WANG, T. Y., LIU, S. J., JONES, K. C., SWEETMAN, A. J. & LU, Y. L. 2013. Industrial source identification and emission estimation of perfluorooctane sulfonate China. *Environment International*, 52, 1-8.
- YAO, T., PU, J., LU, A., WANG, Y. & YU, W. 2007. Recent glacial retreat and its impact on hydrological processes on the Tibetan Plateau, China, and surrounding regions. *Arctic, Antarctic, and Alpine Research*, 39, 642-650.
- YE, Q., ZHU, L., ZHENG, H., NARUSE, R., ZHANG, X. & KANG, S. 2007. Glacier and lake variations in the Yamzhog Yumco basin, southern Tibetan Plateau, from 1980 to 2000 using remote-sensing and GIS technologies. *Journal of Glaciology*, 53, 673-676.
- YOUNG, C. J., FURDUI, V. I., FRANKLIN, J., KOERNER, R. M., MUIR, D. C. & MABURY, S. A. 2007. Perfluorinated acids in arctic snow: new evidence for atmospheric formation. *Environmental Science & Technology*, 41, 3455-3461.
- ZHANG, T., HEGINBOTTOM, J., BARRY, R. G. & BROWN, J. 2000. Further statistics on the distribution of permafrost and ground ice in the Northern Hemisphere 1. *Polar Geography*, 24, 126-131.
- ZHAO, Z., XIE, Z. Y., MOLLER, A., STURM, R., TANG, J. H., ZHANG, G. & EBINGHAUS, R. 2012. Distribution and long-range transport of polyfluoroalkyl substances in the Arctic, Atlantic Ocean and Antarctic coast. *Environmental Pollution*, 170, 71-77.
- ZHU, C., LI, Y., WANG, P., CHEN, Z., REN, D., SSEBUGERE, P., ZHANG, Q. & JIANG, G. 2015. Polychlorinated biphenyls (PCBs) and polybrominated biphenyl ethers (PBDEs) in environmental samples from Ny-Ålesund and London Island, Svalbard, the Arctic. *Chemosphere*, 126, 40-46.

