Chemical hazard in glacial melt? The glacial system as a secondary source of POPs (in the Northern Hemisphere) – a systematic review

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Toxicity of compounds belonging to persistent organic pollutants (POPs) is widely known, and their re-emission from glaciers has been conclusively demonstrated. However, the harmful effects associated with such secondary emissions have yet to be thoroughly understood, especially in the spatial and temporal context, as the existing literature has a clear sampling bias with the best recognition of sites in the European Alps. In this review, we elaborated on the hazards associated with the rapid melting of glaciers releasing organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and polycyclic aromatic hydrocarbons (PAHs). To this end, we collated knowledge on: (1) the varying glacier melt rate across the Northern Hemisphere, (2) the content of POPs in the glacial system components, including the less represented areas, (3) the mechanisms of POPs transfer through the glacial system, including the importance of immediate emission from snow melt, (4) risk assessment associated with POPs re-emission. Based on the limited existing information, the health risk of drinking glacial water can be considered negligible, but consuming aquatic organisms from these waters may increase the risk of cancer. Remoteness from emission sources is a leading factor in the presence of such risk, yet the Arctic is likely to be more exposed to it in the future due to large-scale processes shifting atmospheric pollution and the continuous supply of snow. For future risk monitoring, we recommend to explore the synergistic toxic effects of multiple contaminants and fill the gaps in the spatial distribution of data.

Keywords: OCPs, PCBs, PBDEs, PAHs, glaciers, re-emission, risk

1. Introduction

Glacial ice in the Northern Hemisphere covers a significant part of the Arctic and it occurs at high elevations in Europe, Asia, and North America, with patches in South America and Africa. The contemporary climate change results in rapid glacier melt worldwide (Chandler et al., 2016; Moon, 2017; Zemp et al., 2015). It is also among the drivers of regional water and food security crises, potentially leading to a global crisis if appropriate action is not taken (Fraiture et al., 2010; Hanjra and Qureshi, 2010; Ragab and Prudhomme, 2002). Glacial ice constitutes the largest freshwater reservoir on Earth (National Snow & Ice Data Center, 2020) and its melt impacts multiple environmental components (Miner et al., 2017). Glacier shrinking importantly contributes to the climate-driven water supply vulnerability (IPCC, 2014), especially in the mountain and piedmont regions of the temperate and low latitudes. Approximately 10% of the global population depends partly on glacial water sources (IPCC, 2019). Beside the physical aspect of the water volume supplied by glaciers, the water quality is also of concern (Bogdal et al., 2010; Miner et al., 2017; Wang et al., 2008a).

Meltwater may contain chemical substances deposited on the glacier in the past (Bogdal et al., 2011), including bioaccumulating persistent toxic compounds, both legacy and currently emitted. As chemicals representative of this problem, we chose for the review organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) as legacy anthropogenic persistent pollutants, polycyclic aromatic hydrocarbons (PAHs) as persistent toxic substances of mixed origin, with continuous emissions, and polybrominated diphenyl ethers (PBDEs) as a newer class of compounds with similarly concerning properties (Jones and de Voogt, 1999). Hereafter, we will refer to all those pollutant classes in general as POPs (persistent organic pollutants), despite the PAHs not being listed in the Stockholm Convention (Secretariat of the Stockholm Convention, 2017).

The Arctic and the high mountains are far from direct sources of POPs, yet POPs are detected in the snow, surface water, sediments and animal tissue collected in these regions at disproportionally high concentrations. The “cold condensation” hypothesis (Simonich and Hites, 1995) explains that in temperate climates, volatiles evaporate intensely into the atmosphere, and are transported with warm air masses to be trapped by cooling in polar and alpine regions (Blais et al., 1998; Galassi et al., 1997; Ma et al., 2016).

Since the early 2000s (Blais et al., 2001), it has been conclusively shown that chemicals can occur in discharge from melting glaciers, reintroducing pollution from old emissions. The problem has been studied extensively since, especially in the Alps (Bogdal et al., 2009; Pavlova et al., 2014; Wang et al., 2008a) and Miner et al. (2017) have recently collected the existing literature pertaining directly to this problem in a tutorial review. Miner et al. (2018) have recently investigated the pollution effect in Alaska, an important site for POPs bioaccumulation monitoring due to the fishing industry supporting local communities. Miner et al. (2019, 2018a) provided a risk assessment for pollutants liberated from melting glacier ice and it is the best attempt to date at quantifying the health hazard connected to this secondary wave of pollutant release.

Despite the problem being strongly confirmed in one region of the Northern Hemisphere and present elsewhere, it remains to be determined, in the context of the full pollutant pathway through the glacial system, where such a risk is highest and how it may develop over time. As opposed to the existing tutorial review (Miner et al., 2017) of the problem upon the example of organochlorine legacy POPs, we present here a systematic review on the concentrations of four classes of POPs in the environmental media of the glacial system (for methods of the review see section S1 in Supplementary Information). The following novel insights have been achieved as a result: 1) considering glaciers as secondary sources of both legacy and current emission POPs; 2) collating quantitative data on chemical concentrations of POPs in glacial systems of the Northern Hemisphere, 3) overcoming the spatial sampling bias, which has led to the initial highlighting of 45°N as a maximum pollution zone by Miner et al. (2017), 4) including geographical considerations on the polluted ice distribution and melt timing, 5) renewed review of the possible current and future risk levels connected to the discussed phenomenon.

2. POPs pathways through the glacial system, with an emphasis on the snow cover

Glaciers are complex systems, and the transfer of a POP through them may be simplified in the following way (Fig. 1 & 2). Snowfall, rainfall, and dry deposition are all sources of POPs in glaciers, yet snow is by far the most efficient pathway into the system, both as a scavenger of atmospheric pollutants (Lei and Wania, 2004) and the building material of glaciers. Before the snow is permanently incorporated into the glacier, it may undergo further processes which lead to losses of the pollutant mass stored in them: revolatilisation, decomposition of the pollutant in the snow cover (e.g. through photodegradation; Grannas et al., 2007), and elution with meltwater. It may also capture an extra pollutant load through dry deposition. We focus on processes related to snow cover in this work especially because of their marginal mention in the existing review of the problem (Miner et al., 2017), while other elements of the pollutant pathway are elaborated on in that work.

During snowmelt, the concentrations of various contaminants may change considerably, leading to elevated loadings in some environmental media at the expense of others. A few field (Bizzotto et al., 2009; Kozioł et al., 2017; Lafrenière et al., 2006; Schondorf and Herrmann, 1987; Simmleit et al., 1986) and experimental studies (Meyer et al., 2009a, 2009b; Meyer and Wania, 2011, and refs therein) show that the concentrations of various organic pollutants in natural and artificial snowpack are modified during melt, depending both on the pollutant physicochemical properties and the physical or chemical characteristics of the snowpack. Concentration peaks in meltwater, related to snow and ice melt, have also been studied through modelling (Daly and Wania, 2004; Meyer and Wania, 2011; Morselli et al., 2014). Additionally, Daly and Wania (2004) and Hansen et al. (2006) draw attention to the rapid revolatilisation of the more volatile POPs in the spring, when the snow cover changes properties (grain surface area reduces), resulting in spring emission maxima in the atmosphere, especially for α- and γ-HCH.

A pulse release of the substances accumulated in snow is a potential ecotoxicological hazard (Daly and Wania, 2004), and its impact downstream has been confirmed using a dynamic organism–water–sediment model in the proglacial Frodolfo stream, where 98% of the released mass of PCB-70 and PCB-101 was estimated to come from snow melt, and not ice melt (for p,p'-DDE, on the contrary, 92% came from melting ice; Morselli et al., 2014). In broad terms, the more hydrophilic contaminants (e.g., naphthalene among PAHs) become released from melting snow unproportionally fast, while the longest storage in melting snow is connected to particulate matter affinity. For example, benzo[ghi]perylene is typically released in a pulse at the very end of the snowpack melt, as it binds mainly to particulate impurities in snow (Meyer et al., 2009a). However, such effects are relative: a more dilute, "cleaner" Arctic snowpack will release a higher proportion of its pollution load, even of the more hydrophobic types, at the beginning of the melt, since the initial concentrations are so low that they do not exceed the capacity of water to dissolve these contaminants. On the other hand, snowpacks heavily laden with particulate matter may retain much more of all pollutant types until their last portion melts (Meyer & Wania, 2011). The effect of the elution processes has consequences for the incorporation of POPs into glacial ice in the part of the accumulation area which undergoes significant melt, to the effect of making clean snowpacks even cleaner. Conversely, the particulate-heavy snow would become an even more concentrated reservoir of pollutants. This may be relevant not only to urban snowpacks, but also to glaciers exposed to dust sources (e.g., in Iceland, Kamchatka), although the hypothesis has not been explored yet.

Among snowpack properties, the increasing depth of the snow cover enhances Type 1 elution behavior *sensu* Meyer et al. (2009a), i.e., flushing of more hydrophilic chemicals from the melting snow early; it does not importantly impact any other elution behavior. Particulate matter content and particle permeability of snow regulate the Type 2 (end of melt period) elution. A high internal snow surface area (snow grain surface area) exacerbates Type 3 elution (gradually increasing concentrations in meltwater during the melt season), as opposed to Type 1 which predominates if the grains are coarse and rounded (Meyer & Wania, 2011).

Meyer & Wania (2011) admit to the importance of refreezing in the chemical dynamics of natural snowpacks, yet their model does not elaborate on the effects of this process. A field study of Kozioł et al. (2017) shows that superimposed ice, an effect of basal refreezing, stores the chemicals which would be transported in meltwater. Without the effects of refreezing, the Type 2 (hydrophobic, particle-bound) organic chemicals should dominate the storage in glacial ice. However, refreezing may rebalance the ratios, adding more relatively water-soluble compounds to the mix. Unfortunately, the quantitative impacts of refreezing on the preservation of POPs in glacial ice are unknown to date.

[Figure 1 here]

The snow which has not melted becomes compacted into firn and then ice. The combination of ice flow, glacier shrinkage and upward shift of the boundary between the accumulation and ablation area, i.e. the ELA, leads to the polluted ice being exposed at the glacier front and surface. Meltwater from the polluted layers may be delayed by infiltration towards the glacier bed, yet finally it drains into proglacial rivers. While they typically have a high fine sediment loading, its relatively low organic content may prevent intensive adsorption of POPs onto these particles (Blais et al., 2001). On the other hand, lateral erosion of rivers may incorporate extra pollution load from eroded proglacial soils (Li et al., 2018b). Finally, polluted sediment is deposited either in proglacial lakes, recording former glacial emissions (Bogdal et al., 2010), or in the sea (Pouch et al., 2017). In proglacial waters, the ecosystems process the pollutants, and the top trophic levels of those are especially impacted due to biomagnification (Mazzoni et al., 2020). Both these waters and living organisms in them are subject to human consumption.

[Figure 2 here]

3. Geographical distribution of glacial melt in the context of POPs release

Glaciers and ice sheets worldwide cover around 10% of the land area (IPCC, 2019) and store 69% of global freshwater (National Snow & Ice Data Center, 2020), likely of better water quality than other surface waters. Arguably, contamination within it should draw more focused attention than before. Worldwide, multiple ice masses have been experiencing rapid dwindling, especially the Greenland Ice Sheet (GrIS), with an average mass loss rate of 278 ± 11 Gt yr^{-1}, while all mountain glaciers and ice caps experienced 220 ± 30 Gt yr^{-1} loss (average data for the years 2006-2015; IPCC, 2019).

Glacial mass loss changes in time, which inhibits efficient prediction of POPs reemission in glacial melt. The GrIS provides a striking example, where the loss was exacerbated by 80 ± 6 Gt/y per decade, on average, in the period 1972-2018, reaching –286 ± 20 Gt/y in 2010–2018, although with short-term fluctuations, e.g. due to the cold summers of 2017 and 2018 (Mouginot et al., 2019), or the high melt in the summers of 2012 and 2019 reaching (-600) – (-650) Gt of ice (Velicogna et al., 2020). In GrIS, the majority of mass loss is connected to a small number of outlet glaciers and the coastal area (Forsberg et al., 2017).

Glaciers and ice caps, i.e., all glacial ice outside the GrIS and Antarctic Ice Sheet, follow a different response pattern due to their smaller size and some of them being located in the lower latitudes. In 2006–2016, they have been losing up to 335 ± 144 Gt a^{-1} (Zemp et al., 2019). Future projections (Hock et al., 2019) consistently show at least 22 ± 8% loss of global glacier coverage and 18 ± 7% of volume, including small glaciers and ice caps located in Greenland and Antarctica (and 29 ± 9% and 24 ± 9%, respectively, excluding both), even in the relatively mild RCP2.6 climate scenario. RCP8.5 (“business-as-usual”) will lead to total glacier and ice cap area loss of up to 43 ± 14% and volume loss of up to 36 ± 11% before 2100 (all percentage changes are calculated as means between multiple global glacier mass loss models, and relative to 2015). Temporal distribution of mass losses is also uneven across scenarios: slow to no increase in the absolute ice melt annual rate until 2040 and a decrease afterwards in RCP2.6; as opposed to a constant acceleration of losses (i.e. increasingly negative mass balance) across the 21st century in RCP8.5, with only a plateau towards the 2100 due to many glaciers disappearing (Hock et al., 2019).

To date, the highest cumulative mass losses have been experienced in Western Canada and USA, Central Europe, Alaska, and Asia Central regions (WGMS, 2020) (Fig. 3). In the future, the highest absolute loss is modelled for the regions of Antarctic periphery, Alaska, Arctic Canada North, Greenland periphery, and the Russian Arctic (regardless of the emission scenario), while the relative reductions between 2015 and 2100 would be highest in Central Europe (69 ± 19% for RCP2.6 and 93 ± 10% for RCP 8.5) and lowest in Arctic Canada North (from 12 ± 8% to 23 ± 15% for the respective climate forcings; Hock et al., 2019). Furthermore high percentage losses are projected in Caucasus, Scandinavia, and Low Latitudes; divergent future trajectories could happen in Russian Arctic, Arctic Canada South, Svalbard, and Iceland, especially the latter, which may experience anything between negligible dwindling and an almost total wane of glaciers (Hock et al., 2019).

[Figure 3 here]

Importantly, there are both inherent variability and resulting uncertainty in prediction of the glacier mass balance and its components, different for the summer and the winter balance. Summer balance is typically subject to higher variability and drives the negative total (Medwedeff and Roe, 2017). Furthermore, the same net outcome can be achieved both with high snowfall in winter combined with strong melt in the summer, as with low winter precipitation and slow summer melt. These two situations will have varying impacts on the pollutant storage and release, leading to a fast or slow turnover, respectively. The global climate forcing scenarios RCP2.6, RCP4.5, and RCP8.5 show increasing trends both for temperature and precipitation, with a higher magnitude when the forcing is stronger (Hock et al., 2019; Radić et al., 2014), which could lead to a faster turnover of pollutants and the incorporation of new POPs into the ice. However, the predicted positive temperature trends are stronger in the Arctic (Arctic amplification) (Radić et al., 2014; Shannon et al., 2019), and the forecasted precipitation is much more likely to fall as rain. Under high-end climate scenarios, a substantial drop in snowfall is predicted: by as much as 70% in Central Europe, South Asia east, Svalbard or Iceland. The least affected by such shortages would be the Arctic Canada South, North Asia, and Greenland (-4% – -12%); and only the Arctic Canada North (+22% by 2100) and the Russian Arctic (+7%) would receive extra snowfall then (Shannon et al., 2019).

In the 21st century, models predict a gradual spatial change in glacial melt (maximum meltwater runoff or „peak water"; Huss and Hock, 2018). In the Northern Hemisphere (except most of the Arctic, excluded from the cited study), there are multiple regions with glaciers already past their "peak water" point: most of the European Alps, Canadian Rockies, and the Siberian mountain ranges. Soon (before 2040) peak water will be reached in Norway and the lower parts of the Himalaya. On the other hand, Iceland, Alaska, and the higher parts of the Himalaya are predicted to reach their highest glacial runoff only between 2040 and 2080.

Bliss, Hock and Radić (2014) included all Arctic sites in a similar study, effectively placing Svalbard and Arctic Canada in the same category as Iceland and Alaska (however with a steeper increase before the maximum glacial runoff is reached). They also presented the relationships between runoff trends and the glacier area and terminus elevation, for each region. While the elevation has the strongest impact on the trend direction and magnitude, in most regions, also glacier size plays an important role. Notably, in the context of chemical hazards connected with glacier melt, it appears more dangerous if the maximum melt period does not coincide with the most intensive melt of polluted ice layers, preventing dilution of the supplied pollutant concentrations; the non-uniform distribution of pollutants in the glacial ice requires targeted monitoring to determine the impact of asynchronous pollution and melt peaks. Finally, there occur mechanisms within the glacial system which relocate pollution from the initial contaminant storage, which have yet to be fully understood (see Section 2).

Glaciological understanding offers insights into the location of pollution maxima within the glacier body, as these correspond to ice of certain age. This age can be estimated with the best accuracy at certain points, were ice cores had been drilled, while its spatial distribution can be found through ice flow modeling (in areas with limited data, the accumulation rate provides indirect information). Ice core drilling campaigns have had worldwide coverage. In the Northern Hemisphere, cores have been collected in the Arctic (Greenland, Svalbard, Kamchatka, Alaska, Canadian Arctic) and mountain ranges in the mid-latitudes on all continents (Himalaya and Tibet, the Rockies, European Alps, Tien Shan, Caucasus, and Altai). A multitude of ice dating methods provide cross-validated robust age estimations (Table S1).

The detailed literature review of the investigated ice cores, with respect to the ice thickness corresponding to the maximum emission of the POPs classes of interest in this paper, is provided in Table S1 (Supplementary materials; PAHs are excluded here due to their continuous deposition). We caution that many core studies did not report the newest accumulation due to their early publication dates, so especially PBDE are covered for an uneven number of years. In brief, the measured or calculated layer thickness corresponding to the maximum deposition of PCBs, OCPs, and PBDEs extended between <0.50 and 124 m w.e., depending on the location. Globally, such ice typically spanned the thickest in the European Alps and Alaska, followed closely by Caucasus and Yukon Territory. The thinnest layers corresponded to the maximum POPs accumulation period in Greenland and Ellesmere Island, with Svalbard and the Russian Arctic also in the lower part of the spectrum. At all these locations, the thickness of ice in the period of maximum POPs accumulation rarely exceeded 30 m water equivalent (w.e.), and frequently was below 10 m. However, multiple cores in the Alps showed polluted layer thickness values within that range, too, and the maximum observed in the Alps may encompass higher values than elsewhere due to more studies being conducted there. Nevertheless, the overall picture gathered from point observations indicates important reservoirs of polluted ice remaining in the Alps, Alaska, Yukon, and Caucasus. A full quantitative view would require modelling how far the information from each of those cores can be extrapolated. Indeed, the ice water equivalent related to the legacy pollution period can differ significantly (61-68%) between a single-point measurement and a spatially distributed average, as claimed Pälli et al. (2002) in a study of Svalbard based on radar measurements. An overall picture can be also derived from accumulation rates worldwide, while an important perturbance to such an evaluation is glacier geometry.

The two regions with the best described problem of POPs release from glaciers, i.e., the European Alps and Canadian Rockies, are also those which have already experienced the most rapid melt and have passed their “peak water” (Huss and Hock, 2018). While the Alps may store a further load of these pollutants in their higher parts, other regions with thinner layers of polluted ice are likely to become more important pollutant releasers in the future: at first such locations as Alaska, with a relatively fast ice movement, where the thick layer of polluted ice propagates to the lower parts of the glacier (Hodson, 2014; Miner et al., 2018b). Other regions, where such polluted ice has not been transferred to the ablation zone by ice movement, may join as secondary pollution sources once melting shifts the equilibrium line altitude (ELA) upwards and thus liberates POPs from former accumulation areas. An ELA shifting rapidly and glaciers with a wide tongue (large areas near the current ELA) indicate situations prone to POPs release. Svalbard, Arctic, Canada, Iceland, the Russian Arctic, Greenland periphery, and Himalaya all may yet surprise with locally elevated pollutant release once their thinner layers of POPs storage are exposed. Additionally, Arctic Canada, the Russian Arctic, and Greenland are especially important areas to monitor with respect to emerging contaminants, as they are predicted to receive the most snowfall in the future, which may lead to glaciers in these areas acting as reservoirs for new POPs.

4. Legacy and current use POPs classes accumulated in glaciers

The POPs contain many groups of chemical compounds, and for the capacity of one review we had to narrow our focus to four selected representative classes of POPs. OCPs and PCBs were chosen because they represent old pollutants that had enough time to spread to the most inaccessible parts of the Earth. Their mass use lasted between the end of the Second World War and the 1970s, when restrictions were imposed by individual countries (Breivik et al., 2007; Koureas et al., 2019; Willett et al., 1998). Subsequently, they were banned by the Stockholm Convention (2008), although DDT is still used in some African countries and India to fight malaria (van den Berg et al., 2012). PBDEs are a newer class of impurity, used commercially since late 1960s and surging in 1970s due the limitations in PCB use. By 1990s, PBDEs were widespread (Śmiełowska and Zabiegała, 2018). In the 1990s, penta-BDE were withdrawn in Japan (Jinhui et al., 2017), then in 2004 the EU discontinued penta- and octa-BDE, and by the end of the same year the US decided to ban PBDEs completely. 2008 brought a ban on deca-BDE in the EU (European Court of Justice, 2008) and on all PBDEs in Canada (Jinhui et al., 2017). The Stockholm International Convention adopted in 2009 classified mixtures of penta- and octa-BDE as POPs (Secretariat of the Stockholm Convention, 2011). Deca-BDE restrictions were adopted in 2017, although not as stringent (Secretariat of the Stockholm Convention, 2017). PAHs are formed mainly during incomplete combustion of organic matter, therefore they are of both anthropogenic and natural origin, with continuous production and dispersed sources worldwide (Kozak et al., 2017; Manzetti, 2013; Zhang and Tao, 2009).

5. The contents of POPs in elements of the glacial system

The systematic review below refers to the samples of air, snow, water, and bottom sediments, collected in the vicinity of glaciers, thus representing the glacial system as a whole. Three areas of sampling recurred most frequently: the Alps, the Himalayas, and the Arctic. The review was limited to the Northern Hemisphere due to the extensive data as well as the limited POPs transfer across the equator (UNECE, 2010). We excluded tropical glaciers to avoid treating the equator as a sharp physical boundary between similar glaciers on both hemispheres. Tables S2 and S3, containing concentration ranges, have been placed in Supplementary materials due to their size.

5.1. Atmospheric air

Thanks to the Arctic Monitoring and Assessment Programme (AMAP), the air pollution of POPs in the Arctic is constantly monitored since mid-1990s at four stations: Alert (Ellesmere Island, Canada); Stórhöfði (Iceland); Zeppelin (Ny-Ålesund, Svalbard); and Pallas (Finland). Elsewhere in the Arctic, further short-term research was conducted, including the newer monitoring station in northern Greenland (Bossi et al., 2016). Besides seasonal variability, a downward long-term trend was observed for the atmospheric concentrations of most POPs. However, it has recently been approaching an equilibrium with concentrations in other media (AMAP, 2016). On the Tibetan plateau, long-term monitoring has been carried out since 2008, complemented with a few short-term tests, while in the Alps there are three long-term monitoring stations (Weißfluhjoch Switzerland, Sonnblick Austria and Schneefernerhaus Germany) (Tables S2 &S3).

The existing studies vary by time frame and elevation, which causes a problem in comparing the data as representative for their respective regions. A distinct relationship observed (Fig. 4) is the proportion of volatile compounds increasing with elevation, in accordance with the cold condensation hypothesis (Finizio et al., 2006; Gong et al., 2014).

[Figure 4 here]

(Wang et al., 2019) report that the concentrations of hexachlorobenzene (HCB), α-HCH, γ-HCH, and α-endosulfan are similar in the Arctic and the Tibetan plateau. However, the concentrations shown in their graphs differ from those in the data table (the basis for these graphs). From the data in the original articles, we conclude that higher concentrations of α-HCH and HCB were observed in the Arctic. For the remaining relationships, we found no further differences between the data reported by Wang et al. and the articles they were derived from. In comparison with these two areas, the Alps show the lowest concentration values (except γ-HCH, which are higher). In Nuuk, Greenland, a correlation between γ-HCH and temperature suggested their reemission from soil and snow driving the atmospheric concentrations (Bossi et al., 2008).

5.2. Snow cover

The concentration data reviewed here concern snow samples collected either nearby or on the glaciers themselves. Some publications present concentrations of particular PCB congeners, especially the Dutch 7 (PCB 28, 52, 101, 118, 138, 153, 180) (Table S3), and from these it can be concluded that the highest share in the sum of PCBs (∑PCBs) is brought by the lower chlorine content congeners. These are more volatile and thus prone to travel long distances. Some authors mention also the share of lighter congeners increasing with elevation (Finizio et al., 2006; Kang et al., 2009), which appears in opposition to the claim by Miner et al. (2017) that there are higher transport and deposition rates for organochlorine legacy pollutants with higher molar mass and chlorination (postdepositional processes may produce such a concentration profile instead). Maximum concentrations of ∑PCBs occurred in the Alps and the Arctic, yet due to the large scatter of data, the singular low value from the Himalayas cannot be interpreted as conclusive (Fig. 5a).The highest concentrations of the sum of PAHs (∑PAHs; Fig. 5b), markedly different from the other locations, were observed on Spitsbergen. Such high values may be affected by local emission sources in close proximity to all three sites reported here.

[Figure 5 here]

While we aimed to review the concentration data on 19 selected OCPs, we found sufficient data for comparisons only for HCHs and DDTs. The largest ΣHCHs contents occurred in the Arctic, especially in the Canadian Arctic (Fig. 5c). Those maximum concentrations represent the 1980-90s, when lindane was still widely used. The concentration distribution of ΣDDTs is more even, perhaps due to the greater durability of DDTs compared to HCHs and the continuous use of DDT to combat malaria (Feng et al., 2007). From the collected data pool, which partially overcomes the overrepresentation of samples from the European Alps, it can be concluded that highlighting the latitude of 45°N as a hotspot of pollution by Miner et al. (2017) was premature.

5.3 Ice and firn

Ice cores carry information about the state of the atmosphere in the past, modified through postdepositional changes in the snow cover. Unfortunately, their collection is costly and thus relatively few studies report POPs content in those. To our knowledge, eighteen cores were collected where the content of compounds reviewed here was determined. These ice cores can be divided into two groups, depending on their temporal extent (and the depth they reached).

The first group includes ice cores covering only a few years of accumulation. It consists of cores from Lys Glacier (Villa et al., 2006b), Dasuopu Glacier (Wang et al., 2008c), Mt. Ortles (Kirchgeorg et al., 2016), and Colle Gnifetti (Kirchgeorg et al., 2013). All studies analyzing PCBs and OCPs expose a lack of seasonality in their concentrations. Conversely, for PBDEs, Kirchgeorg et al. (2016) observed a higher concentration in the spring-summer period. There were also differences observed in the case of compounds from the PAHs group, which display a high seasonality associated with the heating period (Wang et al., 2008c).

Ice cores from the second group span several years of accumulation. The highest concentrations of OCPs in those were observed in a few ice cores in the ice layers corresponding to the 1960s-1980s (Donald et al., 1999; Hermanson et al., 2005; Wang et al., 2008a). Maximum concentrations depended on the compound and the latitude of the sampling site, which is related both to the proximity to areas with a longer OCP use period and the long-range atmospheric transport effects (Donald et al., 1999). PCBs and PAHs showed some maxima in concentrations which did not coincide across the hemisphere, yet for PAHs there occurred longer-term increasing trends underlying the short-term variability (Gabrieli et al., 2010; Garmash et al., 2013; Gregor et al., 1995; Kawamura et al., 1994; Pavlova et al., 2015, 2014; Peters et al., 1995; Vecchiato et al., 2020; Vehviläinen et al., 2002). BDE-209 concentrations increased more recently, which matches its emission history (in a core collected prior to PBDE restrictions) (Hermanson et al., 2010). Further temporal considerations on POPs ice core records are discussed in Section 7.2.

5.4 Glacial meltwater

The published studies of the concentrations of OCPs and PCBs in the proglacial streams of the Northern Hemisphere concerned locations in the Alps (Bizzotto et al., 2009; Ferrario et al., 2017; Pavlova et al., 2016; Villa et al., 2006a), Canadian Rockies (Blais et al., 2001; Lafrenière et al., 2006; Miner et al., 2018b), Himalaya (Ali et al., 2018; Sharma et al., 2015) and the Arctic (Kosek et al., 2019, 2018; Mcneely and Gummer, 1984). Although the concentrations of the compounds of interest in stream water were at low levels, their seasonal variability depends on pollutant elution behavior and the availability of its store in glacial ice. Thus, Bizzotto et al. (2009) observed a peak in PCBs concentrations in June, which coincided with the late snowmelt and could be connected to the Type 2 elution behaviour of the higher-chlorinated PCB congeners. The lack of difference between the concentrations in a proglacial and non-glacial stream further confirmed the snowmelt origin of those pollutants in the Frodolfo stream. HCHs, on the other hand, consistently with their more typical elution Type 1 or 3, exhibited high concentrations in May already. They continued to show high concentrations throughout the melt season in the proglacial stream (unlike in the non-glacial one), thus indicating an extra release from glacial ice melt. Additionally HCB and p,p'-DDE exhibited differences in favour of proglacial waters, but only for one month, which could be connected to the early melt of glacial ice. (Villa et al., 2006a) also found higher concentrations of pesticides (especially HCHs and DDTs, less so for HCB) in glacial than in non-glacial waters.

It was also found that the rate of snow melt affects POPs concentrations in streams. Rapid melting causes an increased release of pollutants into the water, while slow melting allows volatiles to escape into the atmosphere instead (Daly and Wania, 2005; Lafrenière et al., 2006). Another modification on the streamwater concentrations of POPs may be the organic content of the sediment it comes in contact with (Blais et al., 2001).

5.5 Proglacial lake sediments

Lake sediments provide information on historical POP emissions, modified by the processes within the upstream part of the catchment. We have found published lake sediment core analyzes of POPs content from the Alps (Bettinetti et al., 2011; Bogdal et al., 2009, 2008; Kohler, M.; Zennegg, M.; Hartmann, P. C.; Sturm, M.; Gujer et al., 2005; Pavlova et al., 2016; Poma et al., 2017; Zennegg et al., 2007), the Himalayas (Guzzella et al., 2011; Sun et al., 2018), and the Arctic (Allen-Gil et al., 1997; Jiao et al., 2009; Mannio et al., 1997; Rose et al., 2004; Sapota et al., 2009; Skotvold and Savinov, 2003). It is impossible to determine an area exposed to the largest amount of pollution, yet there is a common temporal pattern. The maximum content of POPs in sediments occurred during their peak use, then it gradually decreased following the prohibition of use by legal acts. Several publications report a second increase in POP content, signaling their reemission. As this secondary source, melting glaciers are most frequently considered (Bettinetti et al., 2011; Bogdal et al., 2009; Sun et al., 2018), yet lateral erosion of soils by rivers during floods was also proposed (Li et al., 2018a).

5.6 Proglacial soils

Soils are the main place of accumulation of POPs in the terrestrial environment (Kurt-Karakus et al., 2005; Wilcke, 2007). It is believed that the content of clay and natural organic material has a major impact on the ability of soil to accumulate them (Ahn et al., 2006; Boyd et al., 2001; Meijer et al., 2003, 2002), which is consistent with the hydrophobic properties of POPs.

A notable feature is at least ten times higher concentrations of PBDEs in soil in the Alps (Tremolada et al., 2008) than in the Arctic (Wang et al., 2015) and the Tibetan Plateau

(P. Wang et al., 2009; Wang et al., 2012; Zheng et al., 2012). PCBs have also been seen at higher concentrations in the Alps (Tremolada et al., 2008) than in the Tibetan plateau (Zheng et al., 2012). However, no notable differences were observed in the content of OCPs between the Alps (Tremolada et al., 2008) and Tibet (Fu et al., 2001; Tao et al., 2011; Wang et al., 2012; Yuan et al., 2014) and in the content of PAHs between the Arctic (Z. Wang et al., 2009) and Tibet (Tao et al., 2011; Wang et al., 2014, 2013; Yuan et al., 2014).

6. Model understanding of POPs concentrations in the glacial system

A significant progress in understanding glaciers as secondary sources of POPs has been also achieved through modelling. In this section, according to our approach to treat glaciers as a system connected to other environmental media, we will review models of atmospheric transport of POPs and their entrainment into snow cover and glacial ice. However, as other review articles have reported similar findings, we will refer to them and only report the aspects of the state of the art which are the most relevant to glaciers.

6.1. Atmospheric redistribution of POPs into glaciated areas

The atmospheric part of the glacial POPs pathway has been explored through two types of modelling work: 1) the pollution origin interpretation is frequently supported with backward air mass trajectory calculation and 2) the quantitative redistribution of POPs into ice media (including snow) is explored through global or hemispheric POPs fate models. For the first case, HYSPLIT (Rolph et al., 2017; Stein et al., 2015) is frequently used. For example, Hermanson et al. (2020) report that HYSPLIT trajectories show abundant air transport from potential origin areas of organochlorine POPs and Gong et al. (2015) have used HYSPLIT to confirm the long-range transport of OCPs in Tibet. The long-range transport phenomenon highlights the vulnerability of glaciers in remote zones to disproportionally high concentrations of POPs, consistent with the cold condensation hypothesis (Wania and

Mackay, 1995) or the newer, “differential removal hypothesis”, which places more importance with remoteness from POPs sources than with temperature at the receiving location (Friedman and Selin, 2016).

The global redistribution models offer an insight into the future of POPs supply into glaciers hemisphere-wide, especially as their primary emissions diminish. Such models range in complexity (described e.g. by Gusev et al., 2007), and they may also inform about the environmental persistence of various POP types. A thorough review of multiple POPs global fate models for atmospheric applications can be found in the United Nations Hemispheric Transport of Air Pollution 2010 report (UNECE, 2010). In brief, at short time scales, the most important processes governing POPs redistribution are airborne transport, partitioning between gaseous and particulate matter, atmospheric degradation, and deposition. The newer atmospheric models have been refined through the inclusion of more detailed processes (Mu et al., 2017; Whaley et al., 2018). On longer time scales, ocean currents (especially for β-HCH) and reemission from surfaces also contribute importantly (multi-hop mode of transport) (UNECE, 2010). POP fate depends on dry and wet deposition from aerosols and gas, on degradation, volatilization, and physical sequestration (typically into deep sea or soil, although glacial entrainment also belongs in this category). POPs have temperature-dependent physical-chemical properties, which govern their long-range transport.

The behaviour of all POP types mentioned in our review has been subject to atmospheric modelling (OCPs were represented by HCHs and DDT). The evaluation of such models against measurement data shows typical agreement within a factor of three to four or less, yet there occur high discrepancies in some cases, revealing gaps in emission inventory or potential improvements to model structure or parameters. Westerly winds and northward air flow towards the Arctic both transfer significant loads of POPs. For example, PCBs are brought to the Arctic mainly from Europe, Russia, and North America (including through the

Northern Atlantic as a secondary source – Stemmler & Lammel, 2009), and α-HCH from high emission areas in Asia. Emission changes are influential factors in the modification of POPs deposition in receptor regions (which receive up to 30% of POPs from intercontinental transport). These regions would experience a significant drop in POPs deposition load following emission reduction in the main source areas (e.g., Europe for PCBs, North America for B(a)P, or South Asia for α-HCH). In receptor areas, secondary emission sources become more important, for example, they can contribute up to almost 50% of chlorinated POPs in the Arctic; they are much less important in high emission areas like mainland Europe. Atmospheric levels of POPs are influenced by episodic events and thus they need to be equilibrated over longer time periods to estimate their influence on glacial storage. At the time of the UNECE (2010) review, among the main knowledge gaps was listed the description of the impact exerted by secondary sources of POPs on their global fate and long-term trends in source-receptor relationships.

UNECE (2010) reviewed the estimates of climate change impacts on the atmospheric transport of POPs, and these were identified as complex, as they could be exerted through changes in wind fields, atmospheric composition (e.g. aerosol loading), removal efficiency and alterations of the surfaces onto which POPs are deposited, including the shrinking snow and ice cover. The temperature dependence of POPs properties makes them vulnerable to climate change impacts. In a more recent review, focusing especially on climate change impact on the fate of POPs globally, Kallenborn et al. (2012) conclude that the overall effect is obscured by the complex nature of the climate-POP fate relationship and the more important role played by the indirect consequences of climate change (agricultural and industrial). The global shift of activity towards the Arctic combined with the removal of cryospheric barriers and storage (sea ice, snow cover, glaciers) will have a compound effect on POPs contamination. On the other hand, Friedman et al. (2014) showed through their modelling work that for PAHs in the Arctic, that future emission changes will be much more important than secondary effects caused by climate change. Additional concerns are the increased toxicity of pollutants in higher temperatures and the combined effects of climate change and xenobiotics in already vulnerable ecosystems, therefore it is important to determine the tipping points beyond which such effects accelerate (Noyes et al., 2009; Noyes and Lema, 2015).

The issue has been further explored for the Arctic in the ArcRisk project, by using several POPs fate models to estimate both the atmospheric delivery of POPs to the Arctic in the future and their impact on the food web (Carlsson et al., 2018) (with a focus on PCBs, as the ”sentinel” compound group). While the general emission decline will govern the future concentrations of PCBs in the atmosphere, some environmental concentrations in the Arctic may increase and their distribution in the Arctic is likely to be impacted (especially through wet deposition, biological uptake and riverine transport). Remobilisation of PCBs from seawater, soils, and ice is predicted. Carlsson et al. (2018) reviewed PCB fate in the Arctic according to the models BETR Research, MPI-MCTM, DEHM-POP, and ECHAM5-MPIOM. Because the models neglect emission reduction, they have predicted a factor of 2 increase in PCBs concentrations in the Arctic for the ~+3°C climate change scenario. In general, the mid- and high-chlorinated PCBs showed higher sensitivity to climate change (and higher relative concentrations) in the models than did low-chlorinated PCBs. The ECHAM5-MPIOM model included an ocean biogeochemistry submodel, which highlights the increase in bioavailability of PCB-153 due to its efficient binding to colloidal organic matter in the ocean. According to MPI-MCTM, PCB-153 will become more biodegradable, while the persistence of PCB-28 will increase.

The MPI-MCTM was used by Octaviani et al. (2015) also to predict future gateways and transport directions of organochlorine POPs into and out of the Arctic. They concluded that PCB-153 will be net-exported from the Arctic to a higher extent in 2070-2099, while PCB-28 will be net-imported, but at a very low flux level. PCBs will enter the Arctic mainly through Alaska–Northwest Territory, Greenland and Norwegian Sea–Northwest Russia gateways, while exiting the region through the Canadian Arctic, the Denmark Strait and the Russian Far East gateways. DDT will continue to be net-imported into the Arctic, mainly through Alaska–Northwest Territories, Davis Strait–Greenland, Norwegian Sea–Northwestern Russia, and Urals–Siberian gateways. It is noteworthy that an increase in DDT fluxes coming from the Arctic is expected as late as 100 years following their peak emissions. Finally, the authors caution against the effects connected to the newer POPs, giving the example of penta-BDEs, which in the year 2000 have reached a concentration in the Arctic atmosphere which is five times that of PCB153 at its peak primary emission in 1970. Hansen et al. (2015) also predicted that the Arctic will store 38% more HCHs and between 38% less and 17% more PCBs (compared to 1990-2000), due to the redistribution by atmospheric transport. Finally, Friedman and Selin (2016) claim that factors outside the Arctic (whether primary or secondary emissions) will impact the Arctic PCB concentrations stronger in the future than any process happening within the Arctic, thus contradicting the findings of Zhao et al. (2015), who ascribed step changes in Arctic atmospheric PCB concentrations to the effects of a rapid shrinking of sea ice.

A recent review adds information on the atmospheric delivery of PAHs to the Arctic, the main glaciated region of the Northern Hemisphere. Balmer et al. (2019) noted that the fraction of the total environmental burden of three PAHs (anthracene, fluoranthene, and benzo[a]pyrene) stored in the Arctic ranged (between various model scenarios) from 0.5% to 12.8%. Winter and spring in the Canadian Arctic experience orders of magnitude higher airborne concentrations of PAHs, an important seasonal difference favouring their accumulation in glacier ice. Models simulating future changes in atmospheric PAHs concentrations reflect mainly the projected drop in global PAHs emissions, although this effect will be much less pronounced in the Arctic than in the mid-latitudes of the Northern Hemisphere (up to 8% compared to up to 38%).

6.2. Snow and land ice in models

Multi-compartment models at the start of their use did not include separate compartments for the cryosphere. The first to do so were Koziol & Pudykiewicz (2001), who included snow and ice as exchange surfaces in a global distribution study of α- and γ-HCH, yet their model did not allow the analysis of long-term effects, such as storage in glacial ice. A significant improvement in understanding the role of the cryosphere was offered by Hofmann et al. (2012), who included dynamic snow cover and land ice (conceptualized as a stable layer of snow cover over Greenland and Antarctica) in the MPI-MCTM model applied for the global fate of γ-HCH and DDT. In a ten-year simulation, they estimated the recapture in the Northern Hemisphere land ice for 1.83% γ-HCH and 1% DDT. While these numbers may seem small, they corresponded to 20±10% of the total burden of these pollutants in the ice-covered areas. The authors have also estimated the residence times in land ice (4.44 years for DDT and 16.14 years for γ-HCH), although the overall residence times of these substances in the environment were not significantly changed by the inclusion of land ice in the model, because both pollutants are incorporated into the ice mostly at the expense of storage in soil. Similarly, Stocker et al. (2007) have modelled the impact of snow and ice cover as a shield for underlying soil and seawater, finding that the barrier was responsible for between 3% (for αHCH) and 73% (for HCB) decrease in soil concentration against the ice-free world. The seawater in the Arctic was even more influenced by the ice cover, containing only between 8% (for α-HCH) and 21% (for dacthal) of the hypothetical POPs amounts in the ice-free world. The world with the contemporary ice cover produced higher levels of atmospheric pollution (between 2 and 10 times higher, for HCB and PBDE-209, respectively).

The role of snow cover in POPs fate has been investigated by Hansen et al. (2008) in their model DEHM-POP applied to α-HCH behaviour, by comparing the results of model runs with and without snow cover to 21 monitoring datasets. Eight of these sites experienced an improved model fit to reality through the inclusion of the snowpack module, especially predicting spring maxima in air concentrations due to revolatilisation from the snowpack. Another model by Hansen et al. (2006) predicted that α-HCH and γ-HCH are retained in snowpack longer than fluorene, phenanthrene, PCB-28, or PCB-52, which partition more efficiently into the atmospheric air. Finally, the snow scavenging estimation in models may be subject to bias not only from POPs concentration estimations, but also due to the uncertainties in modelling precipitation totals (Whaley et al. 2018).

The specific glacial application of modelling to determine the POPs storage potential has only been applied by one research group. The most extensive models for glaciers as a secondary pollution source have been developed for the Oberaar (Bogdal et al., 2010) and Silvretta glaciers (Steinlin et al., 2016). These studies were able to confirm the role of the glacier as a secondary source of pollution and to determine how much of the deposited pollutant loads has already been released from the Oberaar glacier (half of the PCBs and DDT content being retained in the glacier then) and for the Silvretta glacier. In the latter case, it was estimated that 14 g of PCB-153, 41 g of six indicator PCBs (PCB-28, -52, -101, -138, -153, and -180), and 200 g of ΣPCB was still stored in the glacier. These shares were a small fraction of the PCBs that used to be there. A model of accumulation process in the lake sediment was well adjusted for higher-chlorinated PCB congener content in both studies.

Bogdal et al. (2010) pioneered in explaining englacial fate of POPs with a model and achieved a relatively faithful reproduction of the storage and delay in the system, despite the neglection of transport in meltwater. The reverse order (in time) of pollutant incorporation into the glacial ice and its release was consistent with glaciological understanding. The authors have estimated that due to climate warming, 45% of the stored POPs have been released from Oberaar glacier before 2010, as opposed to 21% in the scenario with stable climate. Furthermore, the changing climate has generated peak emission events beyond the scale expected in the stable scenario.

Steinlin et al. (2016) combined three models to thoroughly describe the fate of PCBs within the glacial system. The models included PCB incorporation into the glacier surface, transport with the ice flow, and chemical fate in the proglacial lake. They were run for the years 1900–2100 and validated against measured PCB concentrations in an ice core, a lake sediment core, and a glacial stream. All PCB fluxes and their storage in the glacier increased until the 1980s and decreased thereafter. The loss dynamics of the lower-chlorinated congeners followed closely the input dynamics, i.e. a high share of these chemicals was released from the glacier immediately after deposition (unlike higher-chlorinated congeners). After a brief increase in the 2000s, the PCB concentrations in the proglacial stream are predicted to be small, although they are expected to persist throughout the 21st century.

Interestingly, ice flowlines explained the high concentrations in the sediment in the 1990s, although not the high concentrations in the 2000s. The proposed explanation for this discrepancy was processes in the ablation area unaccounted for in the model. The proposed delay of PCB delivery into the lake (through, for example, storage in the porous ice layer on the glacier surface) was estimated for 10 years (Steinlin et al., 2016), which could be partly explained by the residence time of the particulate organic matter on the glacier surface as cryoconite (Koziol et al., 2019; Takeuchi et al., 2010). Vertical meltwater percolation within the glacier (not included in the model) could also relocate chemicals (Steinlin et al., 2016). The model predicts a constant concentration of approximately 500 pg L^{-1} ΣPCBs in the glacial stream during the next decades, which is three orders of magnitude lower than the concentration considered chronically toxic for fish; the impacts on higher levels of the trophic chain need further exploration. Steinlin et al. (2016) claim also that the underestimated sorption of lower-chlorinated congeners and the dynamic system of glacial lakes (Nellier et al., 2015) necessitate the inclusion of biochemical processes in the model to explain the accumulation of POPs in lake sediments.

7. Chemical hazards in glacial melt?

OCPs, PCBs and PAHs were found in all the media discussed here (air, snow, ice, stream water, lake sediment). Their presence was also reported by a small number of publications in other media, such as lake water (Diamond et al., 2005; Guzzella et al., 2011; Z. Wang et al., 2009; Yuan et al., 2014) (Tables S2 & S3, Supplementary materials) or seawater (Ademollo et al., 2021) and sea sediment (Pouch et al., 2017). The modest size of the dataset collected so far on these media prevented the interpretation of those for universal spatial and temporal trends. This warrants further studies, especially of the rarely investigated compound classes, such as PBDEs.

7.1. The quality and uniformity of the published data

As the research in this field developed, the methods have been adjusted and as a result the data recorded are not consistent in format across the reviewed works. We describe this here as an important limitation to hemisphere-wide conclusions. A typical problem is differing and non-comparable units, especially of fluxes, e.g., [$\mu g/m^2/y$], and concentrations. In some cases, it was possible to calculate concentrations from fluxes according to standard procedures, including e.g. snow water equivalent, ice density and age, or sediment deposition rate, yet there were instances were unreported raw data hindered our efforts. Some values were available on graphs alone, which decreases the accuracy of reporting. Another difficulty for comparison is the common practice to administer only the sum of the concentrations of compounds for such groups as HCHs or DDTs. In the case of PCBs, the problem is exacerbated because the sum of PCBs may involve a different number of compounds, depending on the study design. For individual PCBs, there are two main ways to present the results: 1) reporting concentrations for 7 indicator PCB compounds (PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153, PCB 180, also named the Dutch seven); 2) grouping PCBs and providing the sums of compound concentrations with the same number of chlorine atoms in the molecule. PAHs experience similar problems as PCBs in reporting the sums. Finally, the concentrations of individual compounds were sometimes given only in the form of an average or a median, which complicated the interpretation, especially as maximum concentrations are of concern for human health impacts.

7.2. The consequences of the pollution trends in time

The rising temperature and enhanced melt release POPs accumulated in glaciers into the environment. This phenomenon has been observed in two ways: in glacial meltwater (through contrasts in concentrations between snow and ice melt and between glacial and non-glacial streams ; Bizzotto et al., 2009) and in sediment cores from proglacial lakes (as delayed secondary concentration maxima; Bettinetti et al., 2011; Bogdal et al., 2009; Sun et al., 2018). Fig. 6 shows the content of ∑DDTs, ∑HCHs, and ∑PCBs in the sediment cores, representing the data for multiple sites in the European Alps and the Himalaya. The release of pollution began at the end of the 1980s in the Himalayas and at the end of the 1990s in the Alps (except Lake Num Co in the Himalaya, where the pollution was also liberated from the 1990s onwards). Within each group of organochlorine legacy compounds, the maximum concentrations from reemission occurred at a similar time. In both regions, the content of OCPs and PCBs in the layers corresponding to reemissions was equal to or even higher than in the layers from the peak use period. In the Alps, concentrations increased steadily until the period of core collection for all POP groups. It was different in the Himalayas: the POP content decreased there in the early 21st century, although it was not uniform for all sites and compounds. Based on these data, POPs re-emission appears to be ongoing. However, there remain important data gaps, especially profiling a wider scope of contaminant concentrations and including areas such as the Arctic, the Rockies, or the rest of Asia.

[Figure 6 here]

The past pollution emission trends are reflected in ice cores, and the layers recorded in those are expected to melt out in reverse order. We have systematically plotted the concentrations of POPs in the existing ice core records to gain new insights into the spatial and temporal distribution of POP-enriched layers (Fig. 7). Considering the concentrations of OCPs in ice cores, it can be stated that most of HCHs and DDTs are accumulated in the European Alpine glaciers, while the Himalaya and the Arctic show lower values. For PCBs, however, there were no clear differences (perhaps the Agassiz ice cap had a higher concentration, but there were Alpine glaciers within a similar range). Therefore, the conclusion of Miner et al. (2017), based on three sites in the Alps and Svalbard, of the much lower concentrations in the Arctic, appears invalid in light of the existing hemisphere-wide data. In most cases, OCPs are accumulated in the layer of their period of maximum use, with the exception of Lys Glacier, where DDT content continued increasing over time. The cause of such a phenomenon, as Villa et al. (2003) explain, was the intensified local emission at the time when DDTs were being phased out. The timing of the PCBs accumulation (and potential release) is more evenly distributed in time, and the distributions in ice cores are at least trimodal. Thus, the PCBs released from melting glaciers may be a longer lasting problem, without a clear peak to trace.

PAHs ice core records are characterized by a different distribution: an underlying slow increase in time (since 1970s), with overlying high extremes of short duration (some overlapping across multiple locations). The slow increase can be linked to the global consumption of fossil fuels and economic growth, while changes in fossil fuel type (especially from coal to petroleum and natural gas) mark drops in these trends (Peters et al., 1995; Vecchiato et al., 2020; Wang et al., 2008b). The short-term maxima may be linked to singular events, such as volcanic eruptions. Especially, there can be found maxima in the hemispheric PAHs ice core record closely coinciding with the eruptions: 1963 Surtsey (Iceland) & 1964 Scheveluch (Kamchatka), 1970 Hekla (Iceland), 1978 Westdhal (Alaska) & 1980 (Mt. St. Helens, Washington, USA), and perhaps also the 1989 Redoubt (Alaska) and 1991 Pinatubo (Philippines). The consequences of such a complex temporal pattern are: difficulties in predicting the exact timing of reemission of PAHs from melting glaciers, regional differences in the impacts of such a phenomenon, and its long duration due to the thicker layer being affected by elevated PAHs concentrations.

[Figure 7 here]

The phenomenon of the reemission of pollutants trapped in a glacier is disturbing, and it shows how legacy pollutants may still cause unexpected problems. It should also be an encouragement to limit the use of any persistent and harmful substances as early as possible, lest they become a similar threat as the DDT. Thus far, the phenomenon has been very well documented in the Alps, observed in the Canadian Rockies, and implied in the Himalayas and Alaska. There have been also concerns raised that due to the rapidly warming climate of the Arctic, the phenomenon will also occur there (Miner et al., 2018b), especially as the cold condensation effect promoted their accumulation in the Arctic climate in the past. As POP sources are more distant in the Arctic and some Arctic glaciers experienced lower past accumulation rates, the amount of pollutants stored in them may be smaller than in mid-latitudes (Hodson, 2014). However, the accumulation phenomenon may be exacerbated in the future due to the transfer of legacy pollution into the Arctic by the global atmospheric circulation (Hansen et al., 2015). While the rest of the Northern Hemisphere is predicted to slowly lose legacy POPs, the Arctic will keep accumulating some of them, especially HCHs.

Newer pollutants may also accumulate faster in the Arctic, according to Hansen et al. (2015). Finally, with the increased human activity in the Arctic, local pollution sources may also increase their impact (e.g., Granberg et al., 2017). Thus, fresh snow monitoring on Arctic glaciers presents an important data source for the future occurrence of the discussed pollution release phenomenon. Especially in the case of PAHs, the measured concentrations in Arctic snow exceed decidedly the concentrations in the Arctic ice cores. Since PAHs emissions are linked to fossil fuel consumption, especially coal combustion, and glacier melt intensity depends on the greenhouse gas emission scenario and dark particle deposition, glacial reemission of POPs could be limited through phasing out fossil fuels, a wider use of particulate filters for combustion, and by using more efficient combustion methods.

7.3. Risk assessment strategies concerning the impact of glacial POPs release

As POPs bioaccumulate (Corsolini and Sarà, 2017; Czub and McLachlan, 2004; Van Drooge et al., 2014; Walters et al., 2008), their contents in fauna and flora have been of particular concern (Bard, 1999; Dietz et al., 2019; Pedersen et al., 2015; Routti et al., 2019). Fortunately, publications on long-term monitoring of the organochlorine POPs content in body tissues from the Arctic report a downward trend in OCP concentrations (a general trend – with exceptions; AMAP, 2017). The reemission from glaciers will impact mainly the organisms found in rivers and lakes to which such meltwater drains, and only in the case of an extremely high and long-term release could this also be observed in the marine ecosystem (Geisz et al., 2008).

The presence of POPs in living organisms, especially in fish, and in drinking water, raises the question of possible harm to human health. Unfortunately, to our knowledge, very few authors have undertaken an assessment of the risk that glacial meltwater poses for human health. Quadroni & Bettinetti (2017) reported that PCB levels in the fish tested sometimes exceeded European Union (EU) standards regarding potential carcinogenic effects. Risk assessment carried out by K. R. Miner et al. (2019) and K.R. Miner et al. (2018) in the Swiss Alps and Alaska, using the Environmental Protection Agency (EPA) methodology, shows that there is no risk associated with the consumption of proglacial water. However, the concentrations in these waters are high enough that the consumption of fish (which bioaccumulate DDTs, HCHs, and PCBs) leads to a risk of cancer. Such risk is greater for children than for adults because of their lower body weight and the expected longer exposure time. At the same time, in waters from another glacier Lys, in the Italian Alps, a lack of risk was concluded for both fish and water consumption, in terms of their DDTs and HCHs concentrations (Miner, 2018). This highlights that the assessed risk level cannot be generalized at a regional scale at the moment, and warrants future research with a wider spatial coverage.

We compared POPs concentrations in glacial streams and rivers with the following water quality guidelines (Table S4): the EPA guidelines, concerning human health (EPA - United States Environmental Protection Agency, 2020), and aquatic life condition (EPA - United States Environmental Protection Agency, 2019) and the Directive of the European Parliament and of the Council on the quality of water intended for human consumption (EU-Groundwater Directive, 2006). The guideline thresholds were only rarely exceeded, among them none of the EU limits. The highest observed concentration in Frodolfo stream (Bizzotto et al., 2009) exceeded the EPA norm more than two times, yet even those concerned a short period of time, while the norm referred to chronic toxicity. Based on these two guidelines, the POPs released from the glacier have not yet caused a known acute or chronic toxicity risk for aquatic organisms or humans.

The aforementioned water quality criteria indicate the risk of cancer associated with the consumption of aquatic organisms and water. Using this criterion, which is based on carcinogenicity risk 10^{-6}, we also scanned the collected database for potential threats. These recommended concentrations of pollutants were exceeded in many cases. The contents of α-HCH (Bizzotto et al., 2009; Ferrario et al., 2017; Lafrenière et al., 2006; Villa et al., 2006a), HCB (Bizzotto et al., 2009), dieldrin (Lafrenière et al., 2006), and p,p'-DDT (Bizzotto et al., 2009; Ferrario et al., 2017; Villa et al., 2006a) were exceeded at a maximum by one order of magnitude. Moreover, PCB (Bizzotto et al., 2009; Ferrario et al., 2017; Lafrenière et al., 2006; Pavlova et al., 2016) and p,p'-DDE (Bizzotto et al., 2009; Ferrario et al., 2017) guideline concentrations were exceeded by almost three orders of magnitude. This fact encourages the continuation of screening for POPs concentrations in glacial meltwater. However, due to the fact that the highest concentrations of PCBs, HCB, and p,p'-DDE in Bizzotto et al. (2009) occurred in the snow melt period (June) and the highest concentrations of α-HCH, dieldrin, and PCBs were noted by Lafrenière et al. (2006) in lysimeters collecting snow melt water, we vouch for including snow elution patterns in the screening study design.

7.4. Recommendations for the chemical monitoring of glaciers

For exposure monitoring, the most important issue to assess is the timing and intensity of the potential pollution plume release from glaciers, based on the limited existing information. With monitoring being both highly demanding in skilled work and logistics, and thus costly, it is not affordable to fund widespread screening programs for POPs in glacial meltwater streams or in other glacier-related media. However, such information is especially needed where the water and fish supply for large groups of people is connected to melting glaciers, and where there are the most vulnerable ecosystems exposed to such extra stressors in the time of an ongoing ecological crisis (Ripple et al., 2019). Thus we seek here for a monitoring strategy robust enough to protect human and animal health.

POPs release from glaciers has been confirmed locally (and regionally in the European Alps), and it is necessary to confirm its occurrence elsewhere, especially since risk levels may differ between glaciers from the same region (Miner, 2018; Miner et al., 2018a). A screening for areas already exposed to POPs release from glaciers (and the chronic toxicity risk) could be based on proglacial lake bottom sediment coring (due to the efficient capture of POPs by sediments). The regions especially vulnerable now and in the future may be located through the interpretation of global chemical fate models, which are increasingly successful at incorporating cryospheric component into the POP cycle, although their spatial distribution could be further refined. Incorporating the functioning of the glacial systems in these models (e.g. ELA shifts, accumulation and ablation rates, glacier geometry) is the next challenge for progress in understanding glaciers as POPs storage.

It is also necessary to determine the combined harm posed by multiple substances released in glacial water. Current models focus mainly on individual chemical compounds or groups of similar compounds, so the risk assessment may be underestimated because it omits other (especially newer) substances and possible synergistic effects. A valuable addition to models would be to use toxicity tests based on living organism exposure (such as in Szczepańska et al., 2017). Due to the risk being mainly connected to either chronic toxicity or carcinogenic effects, any planned monitoring of proglacial waters may be performed with passive samplers exposed for the whole melt season, limiting the number of analyses to be performed. These approaches, combined as befits a particular site, would lead to a more comprehensive assessment of human exposure.

8. Conclusions

The threats connected to the melting of glaciers include multiple physical effects, although the chemicals released from glacial melt can also be dangerous. The increased concentrations of POPs were observed both in sediment cores from proglacial lakes and in glacier-fed streams (as compared to non-glacial streams). Such evidence comes mainly from the European Alps and Canadian Rockies. To overcome this sampling bias, we systematically collected published data on four POP class concentrations in glacial environmental media of the Northern Hemisphere.

While the Canadian Arctic and European Alpine glaciers appear as the most polluted in the hemisphere, and lower pollution concentrations occur in the Himalayas, the regional concentration ranges frequently overlap, spanning <LOD to several ng/L (for some OCPs and the sum of PBDEs). Only the PAHs content in glaciers exceeds that level, and it is clearly the highest in Canadian Arctic at 35-660 ng/L, with intermediate levels in the Caucasus at 49-166 ng/L, the Himalayas at 8.9-100 ng/L or Greenland at 2-103 ng/L, and the lowest in the Alps at 0.5-5.2 ng/L (if locations close to high-emission hotspots are excluded). Based on the existing data, the greatest threats associated with the release of POPs will take place in the Canadian Arctic and the Alps, also due to the presence of thicker ice layers in the maximum emission period. However, multiple regions are too poor in data for a conclusive assessment. Especially, the tentative conclusion of maximum legacy POPs concentrations around 45°N (Miner et al., 2017) is not maintained in light of a wider dataset. Therefore, a more comprehensive way to account for the distance from POPs sources, including local hotspots, and the emissions changes in time needs to be used for spatial assessments (cf. Friedman and Selin, 2016; Kallenborn et al., 2012). Finally, the global pollution shifts and the remaining cryosphere extent will increase the importance of monitoring both legacy and new POPs in the Arctic.

Although the compounds we discuss are toxic, no immediate toxic effects associated with their release in meltwater are to be expected. They may, however, cause harm through long-term effects related to their ability to bioaccumulate and biomagnify. This is unless serious synergic effects are discovered, on which there should be future research, since the overlapping emission periods of various groups of POPs increase such risk. Furthermore, risk assessments are still missing for the release of PAHs and PBDEs in glacial meltwater, including remote locations (Himalaya, Arctic). An excess of PAHs may occur especially in volcanic areas of the Arctic (Iceland, Kamchatka). The northern Canadian Arctic and the Russian Arctic may be continuous recipients of current use pollutants, as they are the only areas predicted to receive extra snowfall in high-end warming climate scenarios. Arctic glaciers and the Himalaya are also important prospective study areas since they are yet to experience their maximum melt later on in the 21st century. Hence, estimating the moment when POPs stored there are released and at what concentrations would identify whether an important chemical hazard to humans or ecosystems will occur.

We suggest applying as monitoring tools for the underrepresented areas: 1) drilling sediment cores from proglacial lakes, 2) monitoring bulk concentrations of POPs in meltwater with passive samplers, 3) employing bioassays to estimate the overall toxicity of proglacial waters. It appears that among legacy pollutants, PCBs will require much longer monitoring due to the lack of clear maxima in their accumulation records in glacier ice (similarly PAHs, due to their continuous emissions). The common emission sources of PAHs and greenhouse gases provides extra arguments for phasing out fossil fuels (especially coal) and applying more efficient combustion methods where possible. For the designation of the most exposed study sites, glaciological parameters can be used: 1) high accumulation rate and accumulation area size at the time of maximum pollutant emission, 2) high melt intensity and a rapid upward ELA shift, 3) the width of the glacier at ELA, which determines the area exposed to melt upon such an upward shift; however, on short-term basis, the role of POPs elution from snow should also be included in monitoring design.

Acknowledgements

F. Pawlak and K. Koziol acknowledge the funding of their research time by the National Science Centre of Poland, project no. NCN 2017/26/D/ST10/00630.

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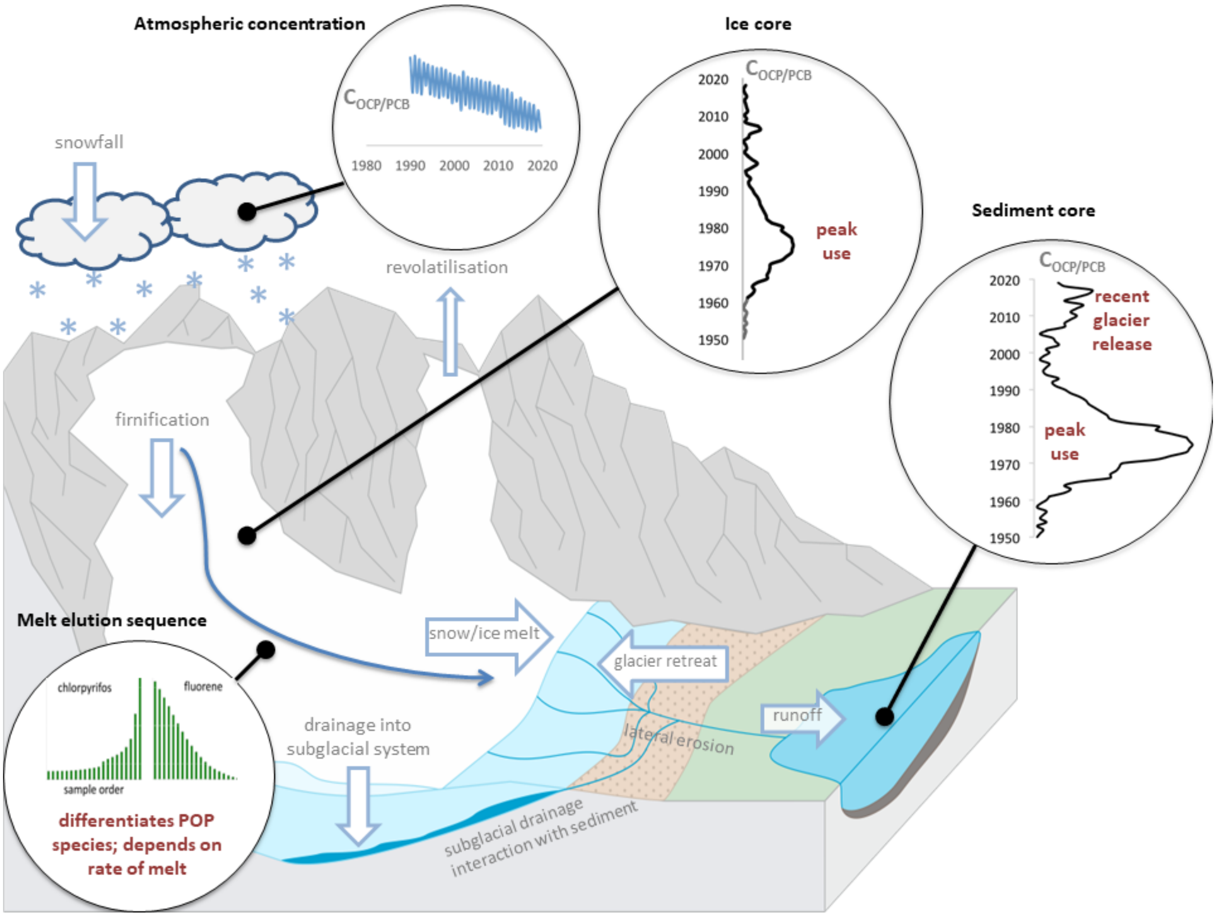


Figure 1. A diagram of the glacial system with the environmental media potentially storing pollutants (with sample pollutant concentration (C_{POP}, $C_{OCP/PCB}$) graphs).

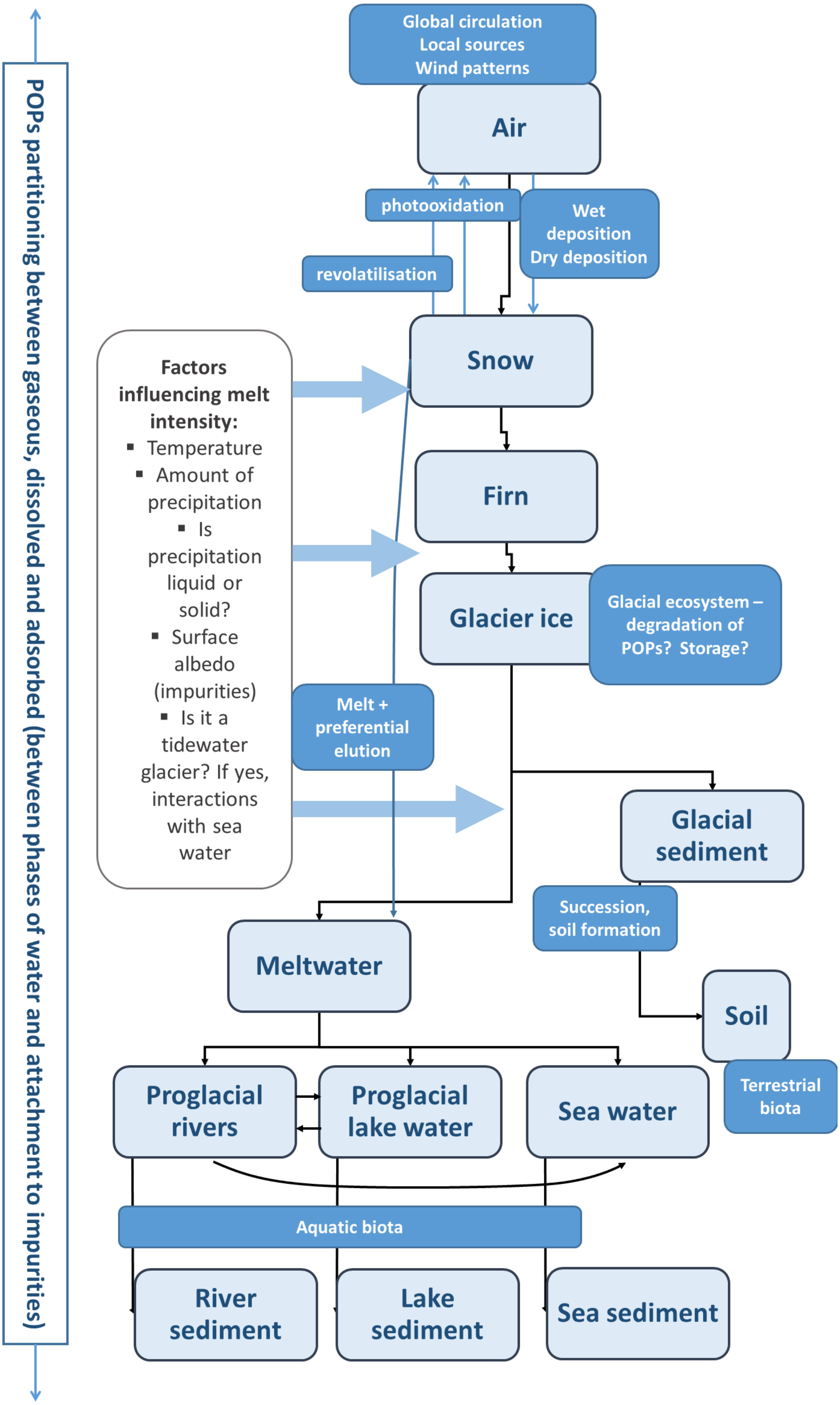

POPs partitioning between gaseous, dissolved and adsorbed (between phases of water and attachment to impurities)
Global circulation
Local sources
Wind patterns
Air
photooxidation
Wet deposition
Dry deposition
revolatilisation
Snow
Factors influencing melt intensity:
▪ Temperature
▪ Amount of precipitation
▪ Is precipitation liquid or solid?
▪ Surface albedo (impurities)
▪ Is it a tidewater glacier? If yes, interactions with sea water
Firn
Glacier ice
Glacial ecosystem – degradation of POPs? Storage?
Melt + preferential elution
Glacial sediment
Succession, soil formation
Soil
Terrestrial biota
Meltwater
Proglacial rivers
Proglacial lake water
Sea water
Aquatic biota
River sediment
Lake sediment
Sea sediment

Figure 2. POPs pathways through the glacial system, including the factors influencing their transport efficiency and storage time.

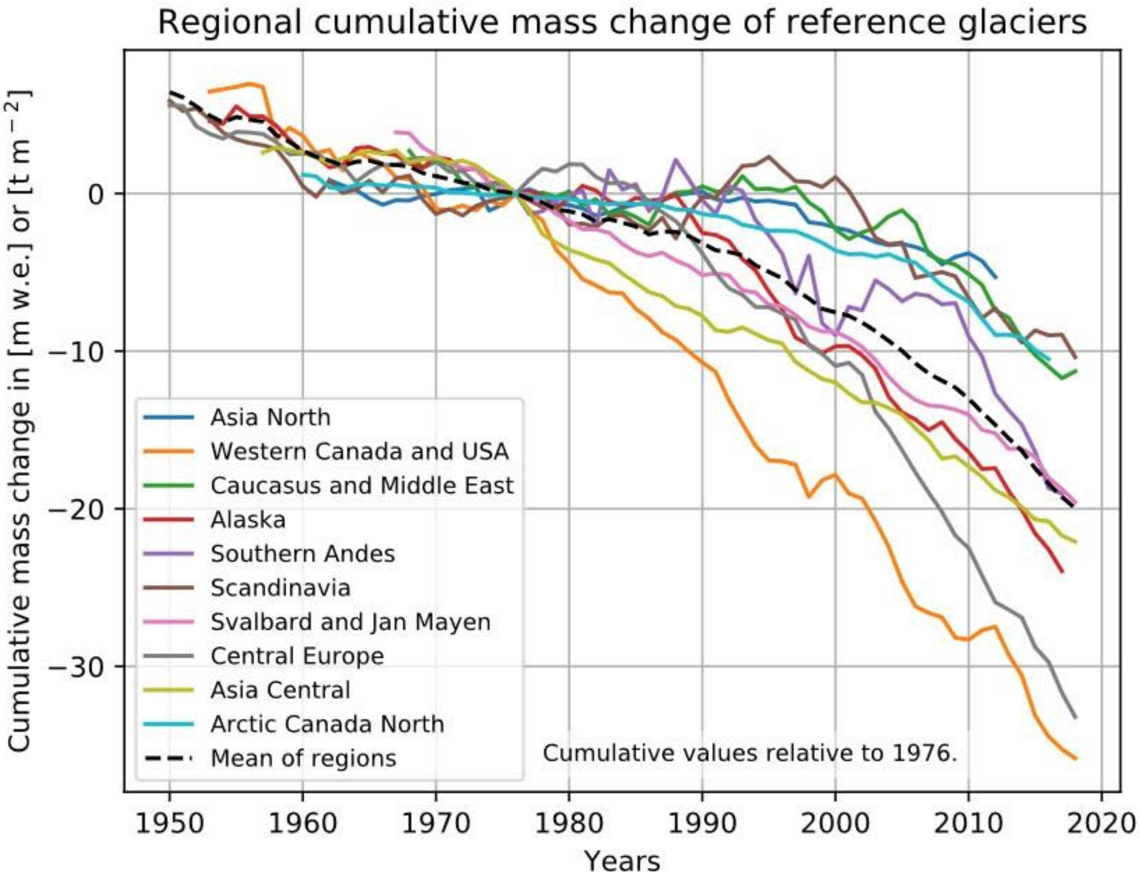


Figure 3. Cumulative mass balance change (since 1976) in glaciers around the world, divided by region. Source: (WGMS, 2020). *Reprinted with permission.*

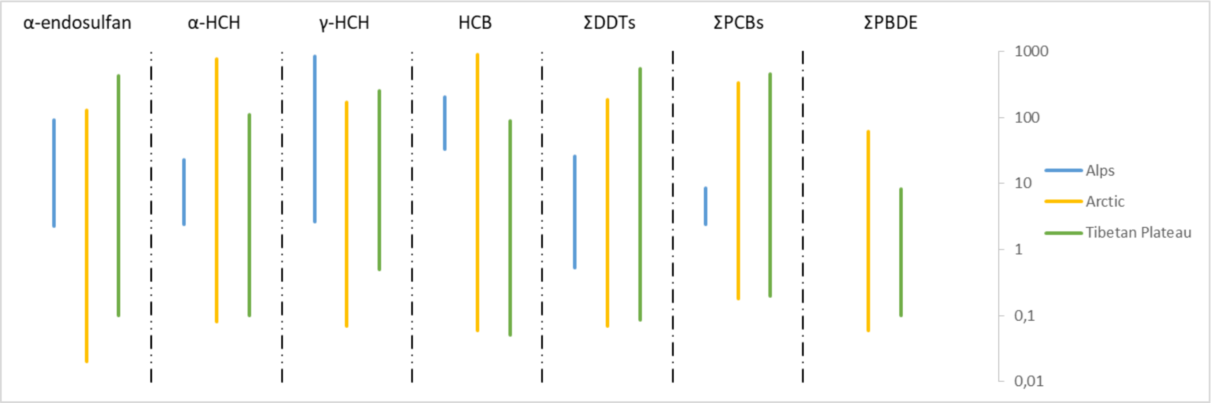
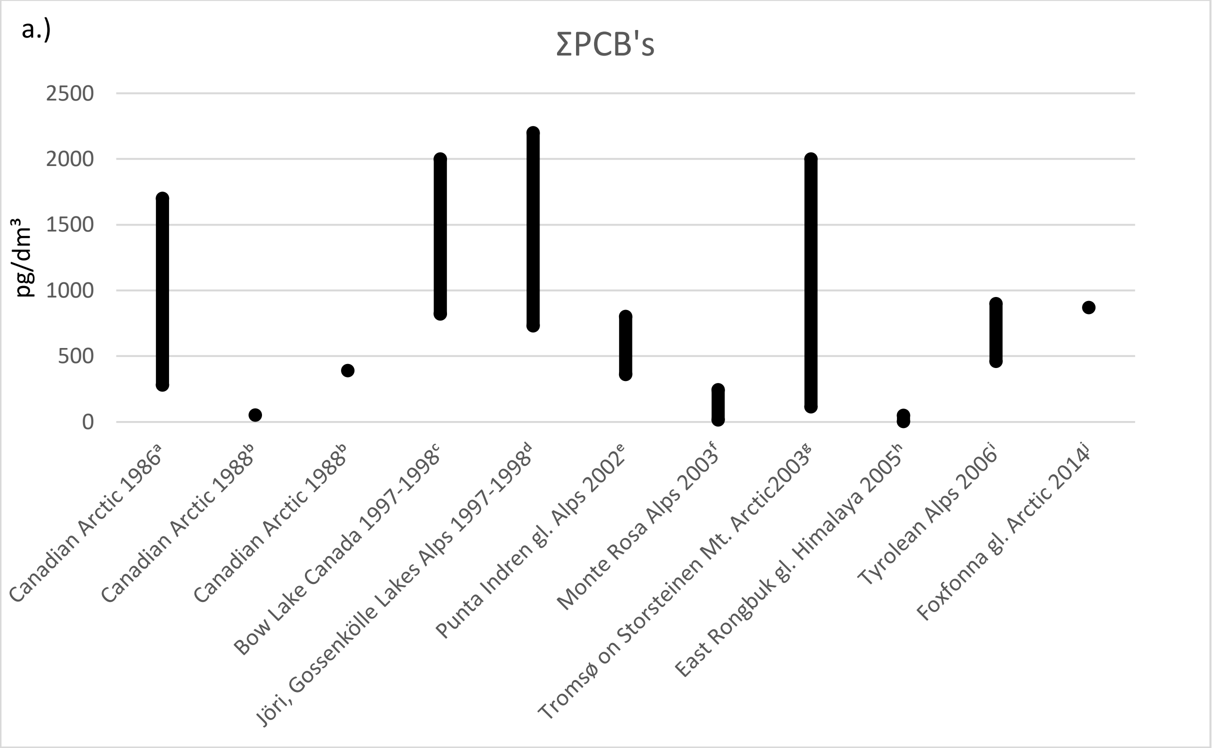
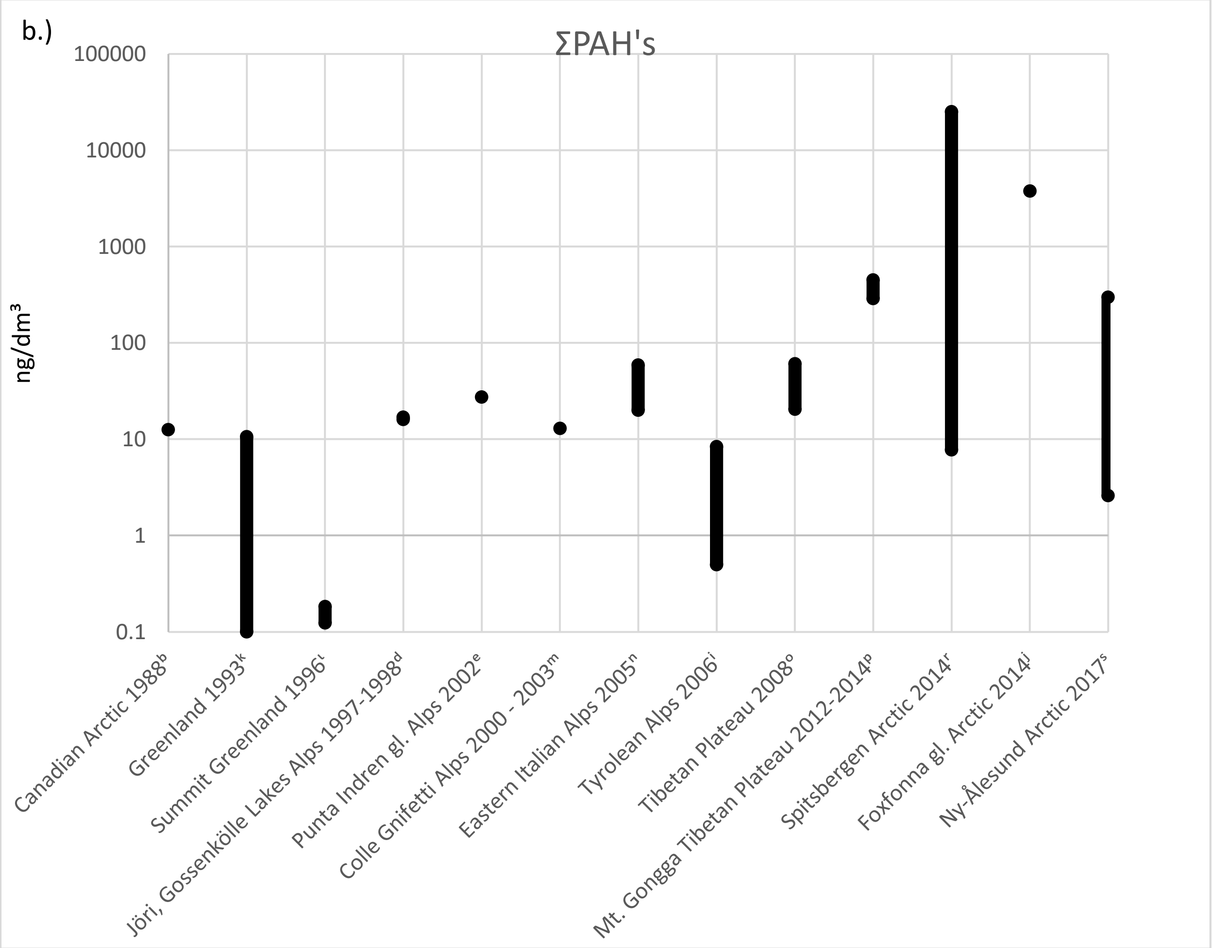


Figure 4. Comparison of the concentration ranges [pg/m³] of atmospheric POPs in the Alps, Arctic and Tibetan Plateau. Data and references used in this figure can be found in Table S2-3.


a.)
ΣPCB's
pg/dm^3
2500
2000
1500
1000
500
0
Canadian Arctic 1986^a
Canadian Arctic 1988^b
Canadian Arctic 1988^b
Bow Lake Canada 1997-1998^c
Jöri, Gossenkölle Lakes Alps 1997-1998^d
Punta Indren gl. Alps 2002^e
Monte Rosa Alps 2003^f
Tromsø on Storsteinen Mt. Arctic2003^g
East Rongbuk gl. Himalaya 2005^h
Tyrolean Alps 2006^i
Foxfonna gl. Arctic 2014^j


b.)
ΣPAH's
ng/dm^3
100000
10000
1000
100
10
1
0.1
Canadian Arctic 1988^b
Greenland 1993^k
Summit Greenland 1996^l
Jöri, Gossenkölle Lakes Alps 1997-1998^d
Punta Indren gl. Alps 2002^e
Colle Gnifetti Alps 2000 - 2003^m
Eastern Italian Alps 2005^n
Tyrolean Alps 2006^i
Tibetan Plateau 2008^o
Mt. Gongga Tibetan Plateau 2012-2014^p
Spitsbergen Arctic 2014^r
Foxfonna gl. Arctic 2014^j
Ny-Ålesund Arctic 2017^s

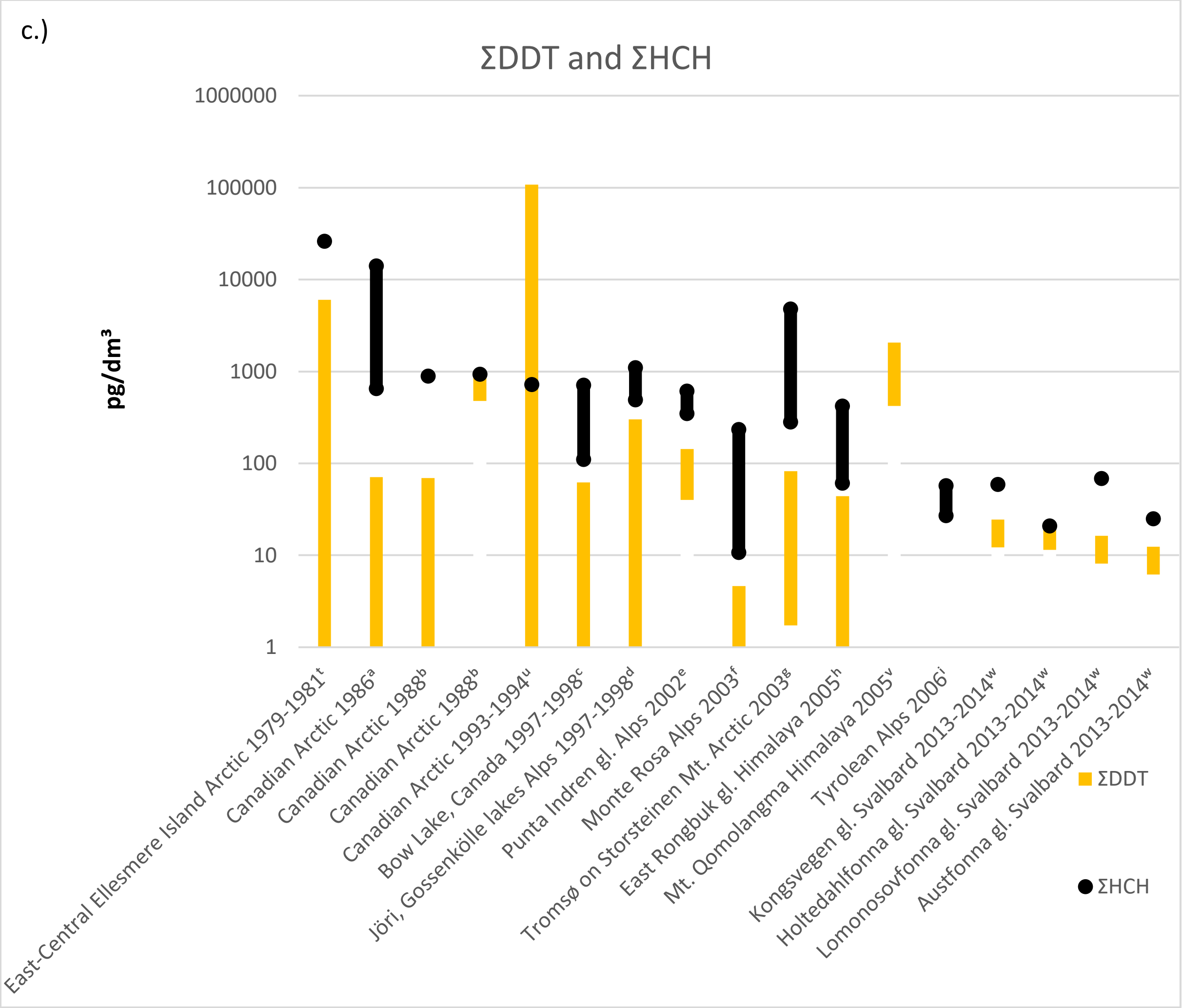
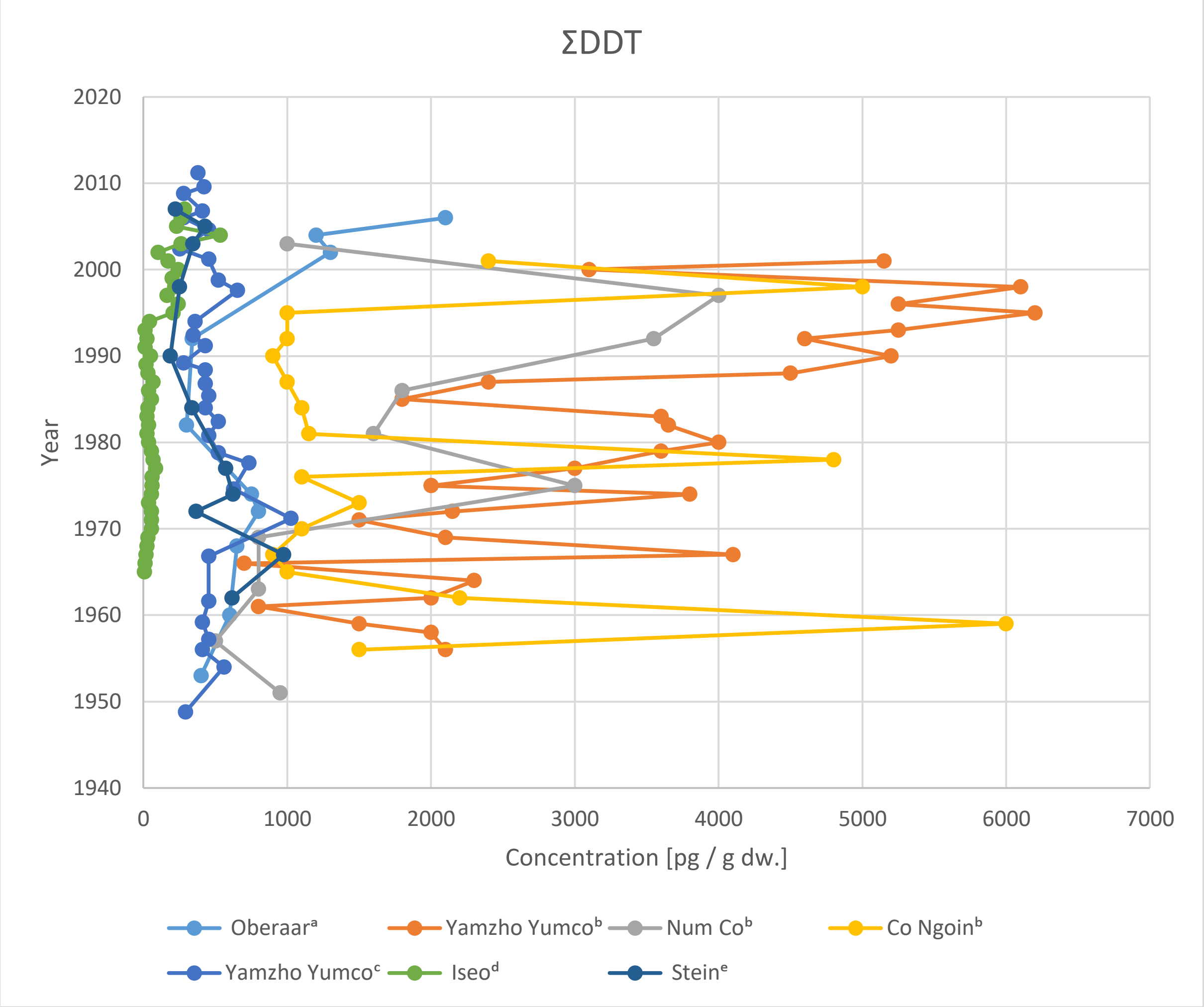
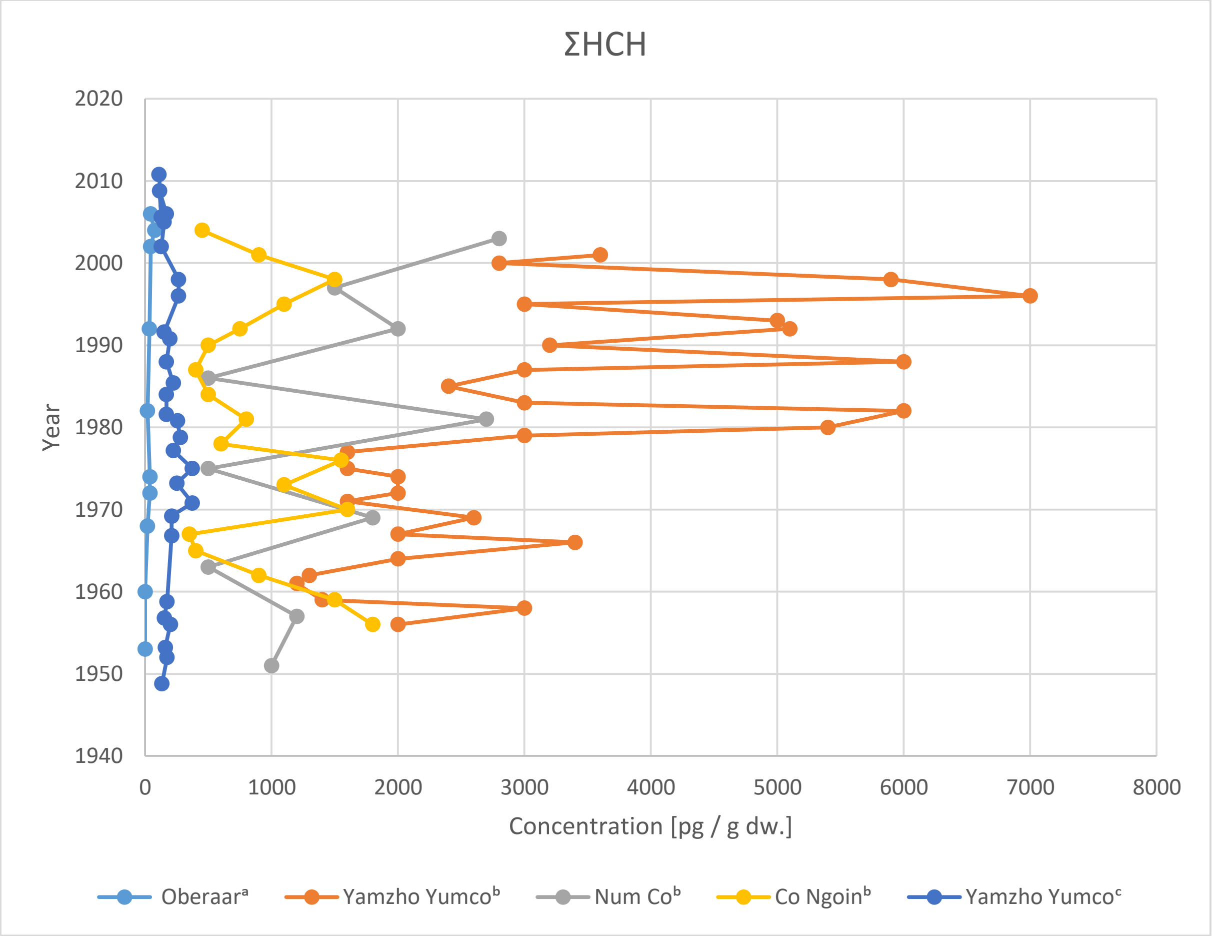


Figure 5. Range of a.) ∑PCBs [pg dm^{-3} of melted snow water], b.) ∑PAHs [ng dm^{-3} of melted snow water] and c.) ∑HCHs and ∑DDTs concentration in [pg dm^{-3} of melted snow water] concentration in snow in various glaciated regions worldwide. a - (Dennis J Gregor & Gummer, 1989), b - (Welch et al., 1991), c - (Lafrenière et al., 2006), d - (Carrera et al., 2001), e - (Herbert et al., 2004), f - (Finizio et al., 2006), g - (Herbert et al., 2005), h - (Kang et al., 2009), i - (Arellano et al., 2014), j - (Kozioł et al., 2017), k – (Masclet et al., 2000), l - (Currie et al., 1998), m - (Gabrieli et al., 2010b)(Gabrieli, Vallelonga, et al., 2010), n - (Gabrieli et al., 2010a)(Gabrieli, Decet, et al., 2010, o - (Li et al., 2011), p - (Yu et al., 2018), r

- (Abramova et al., 2016), s - (Vecchiato et al., 2018), t - (Mcneely and Gummer, 1984), u - (Boyd-Boland et al., 1996), v - (Wang et al., 2007), w - (Hermanson et al., 2020).




ΣHCH
Year
2020
2010
2000
1990
1980
1970
1960
1950
1940
0
1000
2000
3000
4000
5000
6000
7000
8000
Concentration [pg / g dw.]
Oberaarª
Yamzho Yumcoᵇ
Num Coᵇ
Co Ngoinᵇ
Yamzho Yumcoᶜ

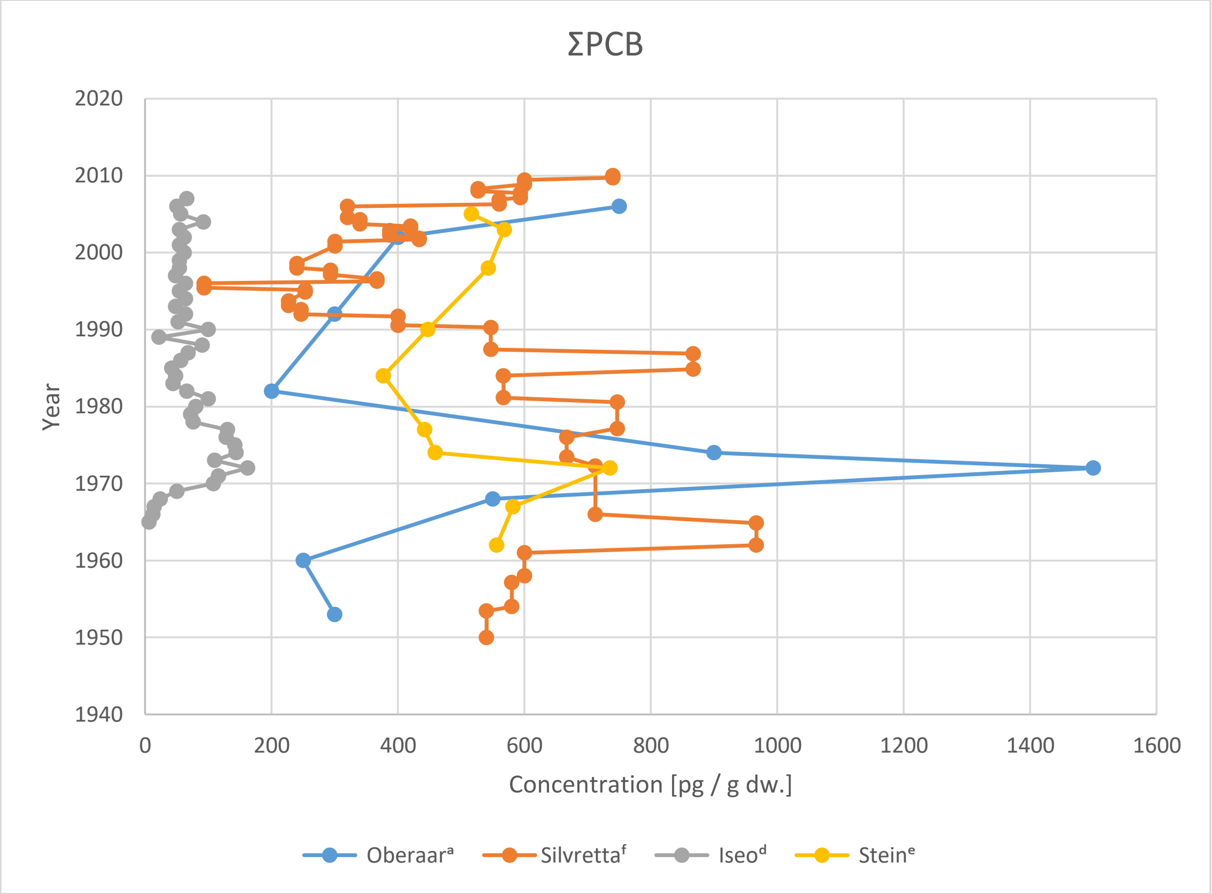
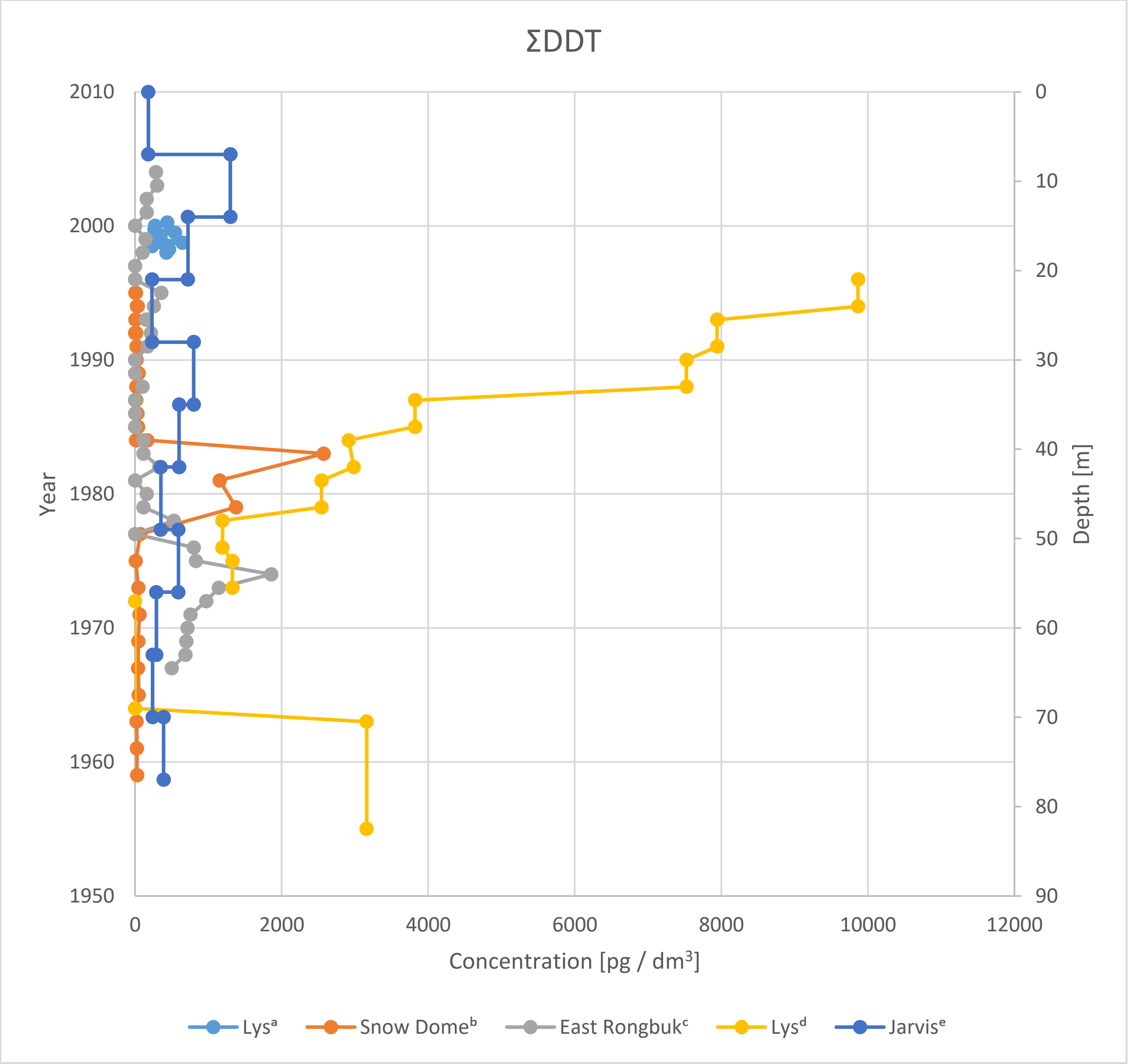
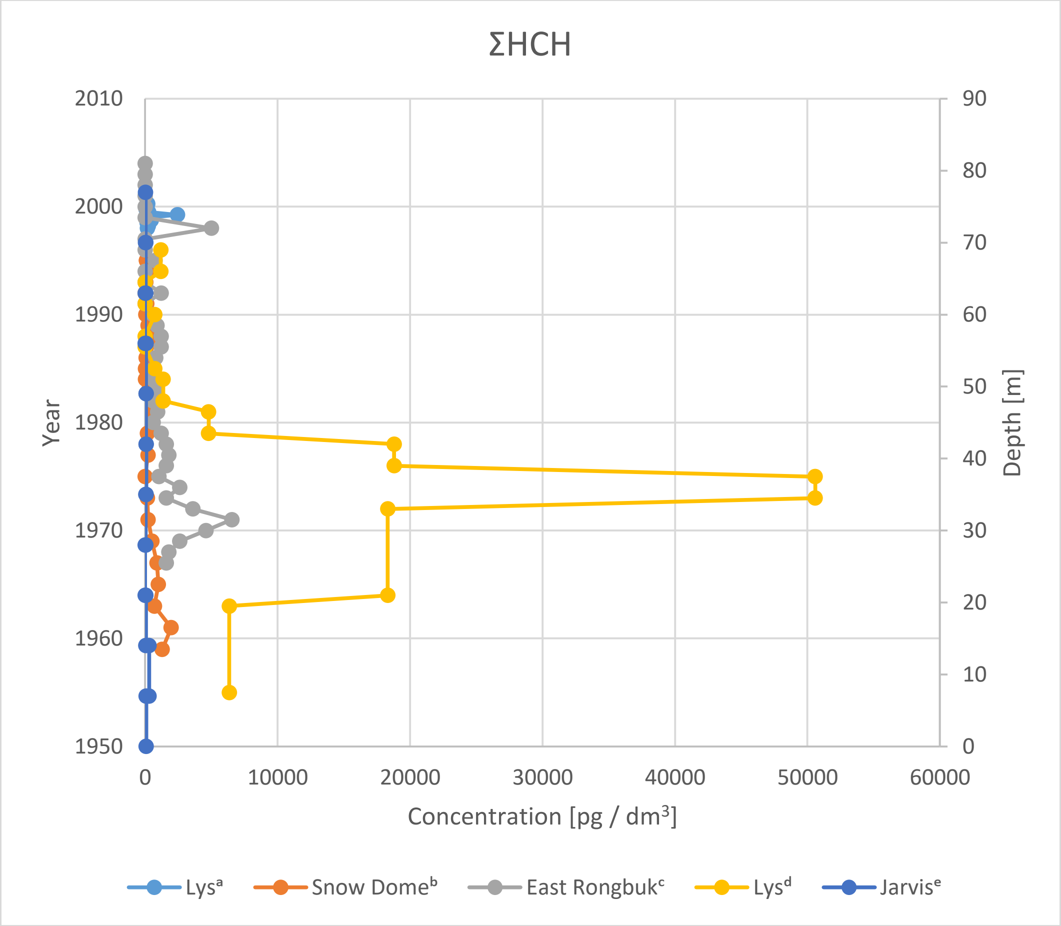
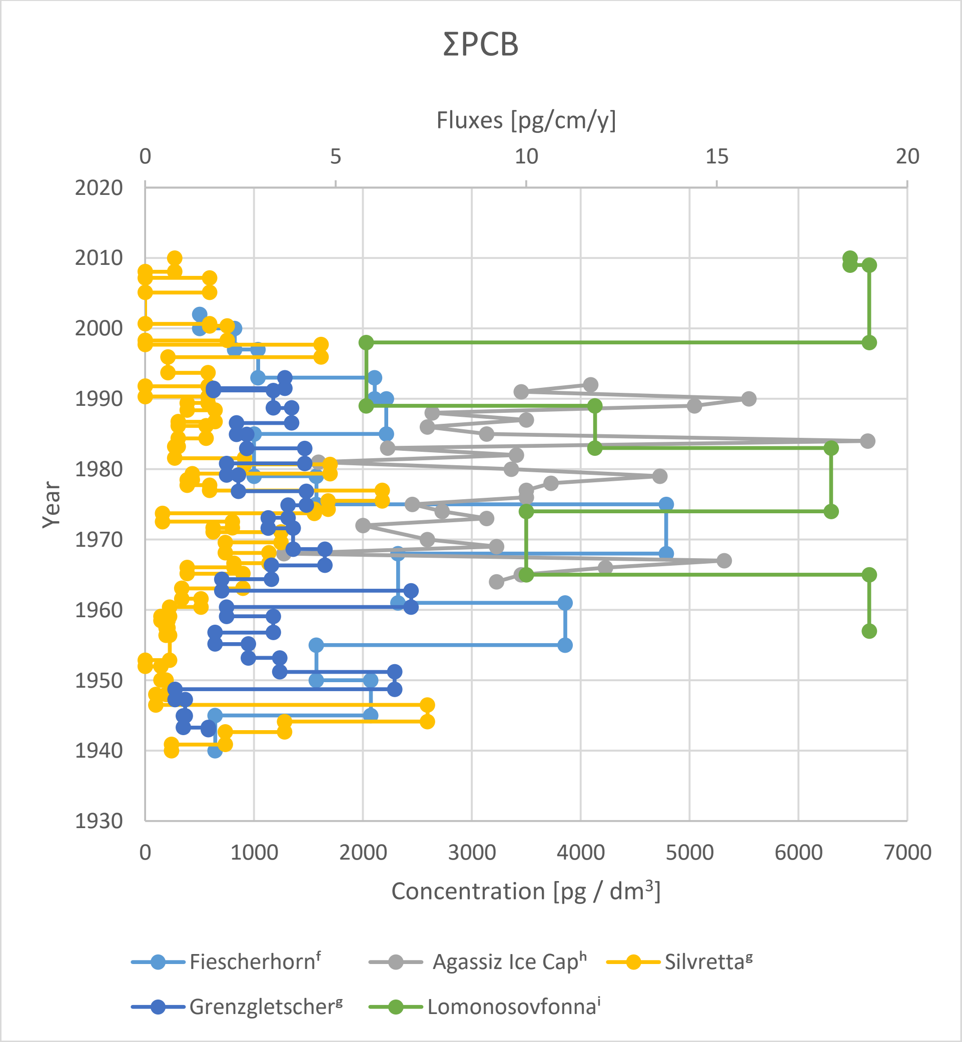


Figure 6. Concentrations of ΣDDT, ΣHCHs, ΣPCBs in the sediment cores from proglacial lakes in the European Alps and Himalaya. a – (Bogdal et al., 2009), b – (Cheng et al., 2014), c – (Sun et al., 2018), d – (Bettinetti et al., 2011), e – (Schmid et al., 2011), f – (Pavlova et al., 2016), e – (Schmid et al., 2011).


ΣDDT
Year
2010
2000
1990
1980
1970
1960
1950
Depth [m]
0
10
20
30
40
50
60
70
80
90
0
2000
4000
6000
8000
10000
12000
Concentration [pg / dm³]
Lys[a]
Snow Dome[b]
East Rongbuk[c]
Lys[d]
Jarvis[e]


ΣHCH
Year
2010
2000
1990
1980
1970
1960
1950
Depth [m]
90
80
70
60
50
40
30
20
10
0
0
10000
20000
30000
40000
50000
60000
Concentration [pg / dm3]
Lys a
Snow Dome b
East Rongbuk c
Lys d
Jarvis e


ΣPCB
Fluxes [pg/cm/y]
0
5
10
15
20
2020
2010
2000
1990
1980
1970
1960
1950
1940
1930
Year
0
1000
2000
3000
4000
5000
6000
7000
Concentration [pg / dm^3]
Fiescherhorn[f]
Agassiz Ice Cap[h]
Silvretta[g]
Grenzgletscher[g]
Lomonosovfonna[i]

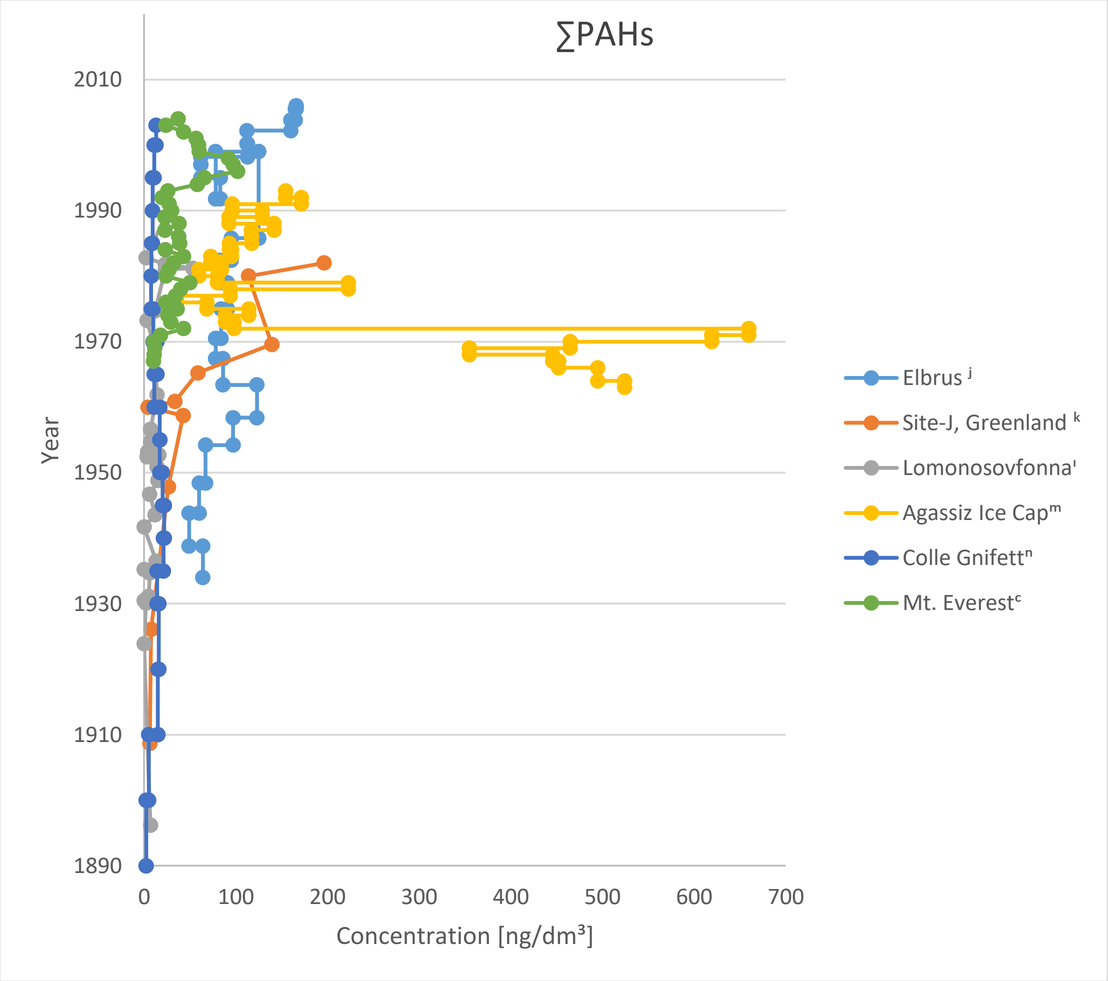


Figure 7. Concentration of ΣDDTs (please note: the age of layers was not specified for Jarvis glacier, hence the concentrations are plotted using depth), ΣHCHs (Jarvis glacier concentrations are plotted by depth instead of age), ΣPCBs (please note that the contents of the ΣPCBs for Lomonosovfonna should be read from the top, secondary axis (as fluxes)), ΣPAHs in the ice cores from published literature. a – (Villa et al., 2006), b – (Donald et al., 1999), c – (Wang et al., 2008), d – (Villa et al., 2003), e - (Miner et al., 2018), f – (P.A. Pavlova et al., 2014), g – (Pavlina Aneva Pavlova et al., 2015), h – (D.J. Gregor et al., 1995), i – (Garmash et al., 2013), j - (Vecchiato et al., 2020), k - (Kawamura et al., 1994), l - (Vehviläinen et al., 2002), m - (Peters et al., 1995), n - (Gabrieli et al., 2010b).

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