

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

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

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

26

27 **1. Introduction**

28 Glacial ice in the Northern Hemisphere covers a significant part of the Arctic and it occurs
29 at high elevations in Europe, Asia, and North America, with patches in South America and
30 Africa. The contemporary climate change results in rapid glacier melt worldwide (Chandler et
31 al., 2016; Moon, 2017; Zemp et al., 2015). It is also among the drivers of regional water and
32 food security crises, potentially leading to a global crisis if appropriate action is not taken
33 (Fraiture et al., 2010; Hanjra and Qureshi, 2010; Ragab and Prudhomme, 2002). Glacial ice
34 constitutes the largest freshwater reservoir on Earth (National Snow & Ice Data Center, 2020)
35 and its melt impacts multiple environmental components (Miner et al., 2017). Glacier
36 shrinking importantly contributes to the climate-driven water supply vulnerability (IPCC,
37 2014), especially in the mountain and piedmont regions of the temperate and low latitudes.
38 Approximately 10% of the global population depends partly on glacial water sources (IPCC,
39 2019). Beside the physical aspect of the water volume supplied by glaciers, the water quality
40 is also of concern (Bogdal et al., 2010; Miner et al., 2017; Wang et al., 2008a).

41 Meltwater may contain chemical substances deposited on the glacier in the past (Bogdal et
42 al., 2011), including bioaccumulating persistent toxic compounds, both legacy and currently
43 emitted. As chemicals representative of this problem, we chose for the review organochlorine
44 pesticides (OCPs) and polychlorinated biphenyls (PCBs) as legacy anthropogenic persistent
45 pollutants, polycyclic aromatic hydrocarbons (PAHs) as persistent toxic substances of mixed
46 origin, with continuous emissions, and polybrominated diphenyl ethers (PBDEs) as a newer
47 class of compounds with similarly concerning properties (Jones and de Voogt, 1999).
48 Hereafter, we will refer to all those pollutant classes in general as POPs (persistent organic

49 pollutants), despite the PAHs not being listed in the Stockholm Convention (Secretariat of the
50 Stockholm Convention, 2017).

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

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

67 Despite the problem being strongly confirmed in one region of the Northern Hemisphere
68 and present elsewhere, it remains to be determined, in the context of the full pollutant
69 pathway through the glacial system, where such a risk is highest and how it may develop over
70 time. As opposed to the existing tutorial review (Miner et al., 2017) of the problem upon the
71 example of organochlorine legacy POPs, we present here a systematic review on the
72 concentrations of four classes of POPs in the environmental media of the glacial system (for
73 methods of the review see section S1 in Supplementary Information). The following novel

74 insights have been achieved as a result: 1) considering glaciers as secondary sources of both
75 legacy and current emission POPs; 2) collating quantitative data on chemical concentrations
76 of POPs in glacial systems of the Northern Hemisphere, 3) overcoming the spatial sampling
77 bias, which has led to the initial highlighting of 45°N as a maximum pollution zone by Miner
78 et al. (2017), 4) including geographical considerations on the polluted ice distribution and
79 melt timing, 5) renewed review of the possible current and future risk levels connected to the
80 discussed phenomenon.

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

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

93 During snowmelt, the concentrations of various contaminants may change considerably,
94 leading to elevated loadings in some environmental media at the expense of others. A few
95 field (Bizzotto et al., 2009; Koziol et al., 2017; Lafrenière et al., 2006; Schondorf and
96 Herrmann, 1987; Simmleit et al., 1986) and experimental studies (Meyer et al., 2009a, 2009b;
97 Meyer and Wania, 2011, and refs therein) show that the concentrations of various organic
98 pollutants in natural and artificial snowpack are modified during melt, depending both on the

99 pollutant physicochemical properties and the physical or chemical characteristics of the
100 snowpack. Concentration peaks in meltwater, related to snow and ice melt, have also been
101 studied through modelling (Daly and Wania, 2004; Meyer and Wania, 2011; Morselli et al.,
102 2014). Additionally, Daly and Wania (2004) and Hansen et al. (2006) draw attention to the
103 rapid revolatilisation of the more volatile POPs in the spring, when the snow cover changes
104 properties (grain surface area reduces), resulting in spring emission maxima in the
105 atmosphere, especially for α - and γ -HCH.

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



124 of pollutants. This may be relevant not only to urban snowpacks, but also to glaciers exposed
125 to dust sources (e.g., in Iceland, Kamchatka), although the hypothesis has not been explored
126 yet.

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

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

142 [Figure 1 here]

143 The snow which has not melted becomes compacted into firn and then ice. The
144 combination of ice flow, glacier shrinkage and upward shift of the boundary between the
145 accumulation and ablation area, i.e. the ELA, leads to the polluted ice being exposed at the
146 glacier front and surface. Meltwater from the polluted layers may be delayed by infiltration
147 towards the glacier bed, yet finally it drains into proglacial rivers. While they typically have a
148 high fine sediment loading, its relatively low organic content may prevent intensive

149 adsorption of POPs onto these particles (Blais et al., 2001). On the other hand, lateral erosion
150 of rivers may incorporate extra pollution load from eroded proglacial soils (Li et al., 2018b).
151 Finally, polluted sediment is deposited either in proglacial lakes, recording former glacial
152 emissions (Bogdal et al., 2010), or in the sea (Pouch et al., 2017). In proglacial waters, the
153 ecosystems process the pollutants, and the top trophic levels of those are especially impacted
154 due to biomagnification (Mazzoni et al., 2020). Both these waters and living organisms in
155 them are subject to human consumption.

156 [Figure 2 here]

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

158 Glaciers and ice sheets worldwide cover around 10% of the land area (IPCC, 2019) and
159 store 69% of global freshwater (National Snow & Ice Data Center, 2020), likely of better
160 water quality than other surface waters. Arguably, contamination within it should draw more
161 focused attention than before. Worldwide, multiple ice masses have been experiencing rapid
162 dwindling, especially the Greenland Ice Sheet (GrIS), with an average mass loss rate of $278 \pm$
163 11 Gt yr^{-1} , while all mountain glaciers and ice caps experienced $220 \pm 30 \text{ Gt yr}^{-1}$ loss
164 (average data for the years 2006-2015; IPCC, 2019).

165 Glacial mass loss changes in time, which inhibits efficient prediction of POPs reemission
166 in glacial melt. The GrIS provides a striking example, where the loss was exacerbated by $80 \pm$
167 6 Gt/y per decade, on average, in the period 1972-2018, reaching $-286 \pm 20 \text{ Gt/y}$ in 2010–
168 2018, although with short-term fluctuations, e.g. due to the cold summers of 2017 and 2018
169 (Mouginot et al., 2019), or the high melt in the summers of 2012 and 2019 reaching (-600) –
170 (-650) Gt of ice (Velicogna et al., 2020). In GrIS, the majority of mass loss is connected to a
171 small number of outlet glaciers and the coastal area (Forsberg et al., 2017).

172 Glaciers and ice caps, i.e., all glacial ice outside the GrIS and Antarctic Ice Sheet, follow a
173 different response pattern due to their smaller size and some of them being located in the



174 lower latitudes. In 2006–2016, they have been losing up to $335 \pm 144 \text{ Gt a}^{-1}$ (Zemp et al.,
175 2019). Future projections (Hock et al., 2019) consistently show at least $22 \pm 8\%$ loss of global
176 glacier coverage and $18 \pm 7\%$ of volume, including small glaciers and ice caps located in
177 Greenland and Antarctica (and $29 \pm 9\%$ and $24 \pm 9\%$, respectively, excluding both), even in
178 the relatively mild RCP2.6 climate scenario. RCP8.5 (“business-as-usual”) will lead to total
179 glacier and ice cap area loss of up to $43 \pm 14\%$ and volume loss of up to $36 \pm 11\%$ before
180 2100 (all percentage changes are calculated as means between multiple global glacier mass
181 loss models, and relative to 2015). Temporal distribution of mass losses is also uneven across
182 scenarios: slow to no increase in the absolute ice melt annual rate until 2040 and a decrease
183 afterwards in RCP2.6; as opposed to a constant acceleration of losses (i.e. increasingly
184 negative mass balance) across the 21st century in RCP8.5, with only a plateau towards the
185 2100 due to many glaciers disappearing (Hock et al., 2019).

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

197 [Figure 3 here]



198 Importantly, there are both inherent variability and resulting uncertainty in prediction of
199 the glacier mass balance and its components, different for the summer and the winter balance.
200 Summer balance is typically subject to higher variability and drives the negative total
201 (Medwedeff and Roe, 2017). Furthermore, the same net outcome can be achieved both with
202 high snowfall in winter combined with strong melt in the summer, as with low winter
203 precipitation and slow summer melt. These two situations will have varying impacts on the
204 pollutant storage and release, leading to a fast or slow turnover, respectively. The global
205 climate forcing scenarios RCP2.6, RCP4.5, and RCP8.5 show increasing trends both for
206 temperature and precipitation, with a higher magnitude when the forcing is stronger (Hock et
207 al., 2019; Radić et al., 2014), which could lead to a faster turnover of pollutants and the
208 incorporation of new POPs into the ice. However, the predicted positive temperature trends
209 are stronger in the Arctic (Arctic amplification) (Radić et al., 2014; Shannon et al., 2019), and
210 the forecasted precipitation is much more likely to fall as rain. Under high-end climate
211 scenarios, a substantial drop in snowfall is predicted: by as much as 70% in Central Europe,
212 South Asia east, Svalbard or Iceland. The least affected by such shortages would be the Arctic
213 Canada South, North Asia, and Greenland (-4% – -12%); and only the Arctic Canada North
214 (+22% by 2100) and the Russian Arctic (+7%) would receive extra snowfall then (Shannon et
215 al., 2019).

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



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

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

245 The detailed literature review of the investigated ice cores, with respect to the ice
246 thickness corresponding to the maximum emission of the POPs classes of interest in this
247 paper, is provided in Table S1 (Supplementary materials; PAHs are excluded here due to their

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

269 The two regions with the best described problem of POPs release from glaciers, i.e., the
270 European Alps and Canadian Rockies, are also those which have already experienced the
271 most rapid melt and have passed their “peak water” (Huss and Hock, 2018). While the Alps
272 may store a further load of these pollutants in their higher parts, other regions with thinner

273 layers of polluted ice are likely to become more important pollutant releasers in the future: at
274 first such locations as Alaska, with a relatively fast ice movement, where the thick layer of
275 polluted ice propagates to the lower parts of the glacier (Hodson, 2014; Miner et al., 2018b).
276 Other regions, where such polluted ice has not been transferred to the ablation zone by ice
277 movement, may join as secondary pollution sources once melting shifts the equilibrium line
278 altitude (ELA) upwards and thus liberates POPs from former accumulation areas. An ELA
279 shifting rapidly and glaciers with a wide tongue (large areas near the current ELA) indicate
280 situations prone to POPs release. Svalbard, Arctic, Canada, Iceland, the Russian Arctic,
281 Greenland periphery, and Himalaya all may yet surprise with locally elevated pollutant
282 release once their thinner layers of POPs storage are exposed. Additionally, Arctic Canada,
283 the Russian Arctic, and Greenland are especially important areas to monitor with respect to
284 emerging contaminants, as they are predicted to receive the most snowfall in the future, which
285 may lead to glaciers in these areas acting as reservoirs for new POPs.

286

287 **4. Legacy and current use POPs classes accumulated in glaciers**

288 The POPs contain many groups of chemical compounds, and for the capacity of one
289 review we had to narrow our focus to four selected representative classes of POPs. OCPs and
290 PCBs were chosen because they represent old pollutants that had enough time to spread to the
291 most inaccessible parts of the Earth. Their mass use lasted between the end of the Second
292 World War and the 1970s, when restrictions were imposed by individual countries (Breivik et
293 al., 2007; Koureas et al., 2019; Willett et al., 1998). Subsequently, they were banned by the
294 Stockholm Convention (2008), although DDT is still used in some African countries and
295 India to fight malaria (van den Berg et al., 2012). PBDEs are a newer class of impurity, used
296 commercially since late 1960s and surging in 1970s due the limitations in PCB use. By 1990s,
297 PBDEs were widespread (Śmiełowska and Zabiegała, 2018). In the 1990s, penta-BDE were



298 withdrawn in Japan (Jinhui et al., 2017), then in 2004 the EU discontinued penta- and octa-
299 BDE, and by the end of the same year the US decided to ban PBDEs completely. 2008
300 brought a ban on deca-BDE in the EU (European Court of Justice, 2008) and on all PBDEs in
301 Canada (Jinhui et al., 2017). The Stockholm International Convention adopted in 2009
302 classified mixtures of penta- and octa-BDE as POPs (Secretariat of the Stockholm
303 Convention, 2011). Deca-BDE restrictions were adopted in 2017, although not as stringent
304 (Secretariat of the Stockholm Convention, 2017). PAHs are formed mainly during incomplete
305 combustion of organic matter, therefore they are of both anthropogenic and natural origin,
306 with continuous production and dispersed sources worldwide (Kozak et al., 2017; Manzetti,
307 2013; Zhang and Tao, 2009).

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

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

317 **5.1. Atmospheric air**

318 Thanks to the Arctic Monitoring and Assessment Programme (AMAP), the air
319 pollution of POPs in the Arctic is constantly monitored since mid-1990s at four stations: Alert
320 (Ellesmere Island, Canada); Stórhöfði (Iceland); Zeppelin (Ny-Ålesund, Svalbard); and Pallas
321 (Finland). Elsewhere in the Arctic, further short-term research was conducted, including the
322 newer monitoring station in northern Greenland (Bossi et al., 2016). Besides seasonal



323 variability, a downward long-term trend was observed for the atmospheric concentrations of
324 most POPs. However, it has recently been approaching an equilibrium with concentrations in
325 other media (AMAP, 2016). On the Tibetan plateau, long-term monitoring has been carried
326 out since 2008, complemented with a few short-term tests, while in the Alps there are three
327 long-term monitoring stations (Weißfluhjoch Switzerland, Sonnblick Austria and
328 Schneefernerhaus Germany) (Tables S2 &S3).

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

333 [Figure 4 here]

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

344 **5.2. Snow cover**

345 The concentration data reviewed here concern snow samples collected either nearby or
346 on the glaciers themselves. Some publications present concentrations of particular PCB

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

360 [Figure 5 here]

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

370 ***5.3 Ice and firn***

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

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

384 Ice cores from the second group span several years of accumulation. The highest
385 concentrations of OCPs in those were observed in a few ice cores in the ice layers
386 corresponding to the 1960s-1980s (Donald et al., 1999; Hermanson et al., 2005; Wang et al.,
387 2008a). Maximum concentrations depended on the compound and the latitude of the sampling
388 site, which is related both to the proximity to areas with a longer OCP use period and the
389 long-range atmospheric transport effects (Donald et al., 1999). PCBs and PAHs showed some
390 maxima in concentrations which did not coincide across the hemisphere, yet for PAHs there
391 occurred longer-term increasing trends underlying the short-term variability (Gabrieli et al.,
392 2010; Garmash et al., 2013; Gregor et al., 1995; Kawamura et al., 1994; Pavlova et al., 2015,
393 2014; Peters et al., 1995; Vecchiato et al., 2020; Vehviläinen et al., 2002). BDE-209
394 concentrations increased more recently, which matches its emission history (in a core

395 collected prior to PBDE restrictions) (Hermanson et al., 2010). Further temporal
396 considerations on POPs ice core records are discussed in Section 7.2.

397 *5.4 Glacial meltwater*

398 The published studies of the concentrations of OCPs and PCBs in the proglacial
399 streams of the Northern Hemisphere concerned locations in the Alps (Bizzotto et al., 2009;
400 Ferrario et al., 2017; Pavlova et al., 2016; Villa et al., 2006a), Canadian Rockies (Blais et al.,
401 2001; Lafrenière et al., 2006; Miner et al., 2018b), Himalaya (Ali et al., 2018; Sharma et al.,
402 2015) and the Arctic (Kosek et al., 2019, 2018; Mcneely and Gummer, 1984). Although the
403 concentrations of the compounds of interest in stream water were at low levels, their seasonal
404 variability depends on pollutant elution behavior and the availability of its store in glacial ice.
405 Thus, Bizzotto et al. (2009) observed a peak in PCBs concentrations in June, which coincided
406 with the late snowmelt and could be connected to the Type 2 elution behaviour of the higher-
407 chlorinated PCB congeners. The lack of difference between the concentrations in a proglacial
408 and non-glacial stream further confirmed the snowmelt origin of those pollutants in the
409 Frodolfo stream. HCHs, on the other hand, consistently with their more typical elution Type 1
410 or 3, exhibited high concentrations in May already. They continued to show high
411 concentrations throughout the melt season in the proglacial stream (unlike in the non-glacial
412 one), thus indicating an extra release from glacial ice melt. Additionally HCB and p,p'-DDE
413 exhibited differences in favour of proglacial waters, but only for one month, which could be
414 connected to the early melt of glacial ice. (Villa et al., 2006a) also found higher
415 concentrations of pesticides (especially HCHs and DDTs, less so for HCB) in glacial than in
416 non-glacial waters.

417 It was also found that the rate of snow melt affects POPs concentrations in streams.
418 Rapid melting causes an increased release of pollutants into the water, while slow melting
419 allows volatiles to escape into the atmosphere instead (Daly and Wania, 2005; Lafrenière et

420 al., 2006). Another modification on the streamwater concentrations of POPs may be the
421 organic content of the sediment it comes in contact with (Blais et al., 2001).

422 ***5.5 Proglacial lake sediments***

423 Lake sediments provide information on historical POP emissions, modified by the
424 processes within the upstream part of the catchment. We have found published lake sediment
425 core analyzes of POPs content from the Alps (Bettinetti et al., 2011; Bogdal et al., 2009,
426 2008; Kohler, M.; Zennegg, M.; Hartmann, P. C.; Sturm, M.; Gujer et al., 2005; Pavlova et
427 al., 2016; Poma et al., 2017; Zennegg et al., 2007), the Himalayas (Guzzella et al., 2011; Sun
428 et al., 2018), and the Arctic (Allen-Gil et al., 1997; Jiao et al., 2009; Mannio et al., 1997; Rose
429 et al., 2004; Sapota et al., 2009; Skotvold and Savinov, 2003). It is impossible to determine an
430 area exposed to the largest amount of pollution, yet there is a common temporal pattern. The
431 maximum content of POPs in sediments occurred during their peak use, then it gradually
432 decreased following the prohibition of use by legal acts. Several publications report a second
433 increase in POP content, signaling their reemission. As this secondary source, melting glaciers
434 are most frequently considered (Bettinetti et al., 2011; Bogdal et al., 2009; Sun et al., 2018),
435 yet lateral erosion of soils by rivers during floods was also proposed (Li et al., 2018a).

436 ***5.6 Proglacial soils***

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

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

444 (P. Wang et al., 2009; Wang et al., 2012; Zheng et al., 2012). PCBs have also been seen at
445 higher concentrations in the Alps (Tremolada et al., 2008) than in the Tibetan plateau (Zheng
446 et al., 2012). However, no notable differences were observed in the content of OCPs between
447 the Alps (Tremolada et al., 2008) and Tibet (Fu et al., 2001; Tao et al., 2011; Wang et al.,
448 2012; Yuan et al., 2014) and in the content of PAHs between the Arctic (Z. Wang et al., 2009)
449 and Tibet (Tao et al., 2011; Wang et al., 2014, 2013; Yuan et al., 2014).

450 **6. Model understanding of POPs concentrations in the glacial system**

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

457 ***6.1. Atmospheric redistribution of POPs into glaciated areas***

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

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

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

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

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

506 UNECE (2010) reviewed the estimates of climate change impacts on the atmospheric
507 transport of POPs, and these were identified as complex, as they could be exerted through
508 changes in wind fields, atmospheric composition (e.g. aerosol loading), removal efficiency
509 and alterations of the surfaces onto which POPs are deposited, including the shrinking snow
510 and ice cover. The temperature dependence of POPs properties makes them vulnerable to
511 climate change impacts. In a more recent review, focusing especially on climate change
512 impact on the fate of POPs globally, Kallenborn et al. (2012) conclude that the overall effect
513 is obscured by the complex nature of the climate-POP fate relationship and the more
514 important role played by the indirect consequences of climate change (agricultural and
515 industrial). The global shift of activity towards the Arctic combined with the removal of
516 cryospheric barriers and storage (sea ice, snow cover, glaciers) will have a compound effect
517 on POPs contamination. On the other hand, Friedman et al. (2014) showed through their

518 modelling work that for PAHs in the Arctic, that future emission changes will be much more
519 important than secondary effects caused by climate change. Additional concerns are the
520 increased toxicity of pollutants in higher temperatures and the combined effects of climate
521 change and xenobiotics in already vulnerable ecosystems, therefore it is important to
522 determine the tipping points beyond which such effects accelerate (Noyes et al., 2009; Noyes
523 and Lema, 2015).

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

541 The MPI-MCTM was used by Octaviani et al. (2015) also to predict future gateways
542 and transport directions of organochlorine POPs into and out of the Arctic. They concluded



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

561 A recent review adds information on the atmospheric delivery of PAHs to the Arctic, the main
562 glaciated region of the Northern Hemisphere. Balmer et al. (2019) noted that the fraction of
563 the total environmental burden of three PAHs (anthracene, fluoranthene, and benzo[a]pyrene)
564 stored in the Arctic ranged (between various model scenarios) from 0.5% to 12.8%. Winter
565 and spring in the Canadian Arctic experience orders of magnitude higher airborne
566 concentrations of PAHs, an important seasonal difference favouring their accumulation in
567 glacier ice. Models simulating future changes in atmospheric PAHs concentrations reflect

568 mainly the projected drop in global PAHs emissions, although this effect will be much less
569 pronounced in the Arctic than in the mid-latitudes of the Northern Hemisphere (up to 8%
570 compared to up to 38%).

571 ***6.2. Snow and land ice in models***

572 Multi-compartment models at the start of their use did not include separate
573 compartments for the cryosphere. The first to do so were Koziol & Pudykiewicz (2001), who
574 included snow and ice as exchange surfaces in a global distribution study of α - and γ -HCH,
575 yet their model did not allow the analysis of long-term effects, such as storage in glacial ice.
576 A significant improvement in understanding the role of the cryosphere was offered by
577 Hofmann et al. (2012), who included dynamic snow cover and land ice (conceptualized as a
578 stable layer of snow cover over Greenland and Antarctica) in the MPI-MCTM model applied
579 for the global fate of γ -HCH and DDT. In a ten-year simulation, they estimated the recapture
580 in the Northern Hemisphere land ice for 1.83% γ -HCH and 1% DDT. While these numbers
581 may seem small, they corresponded to $20 \pm 10\%$ of the total burden of these pollutants in the
582 ice-covered areas. The authors have also estimated the residence times in land ice (4.44 years
583 for DDT and 16.14 years for γ -HCH), although the overall residence times of these substances
584 in the environment were not significantly changed by the inclusion of land ice in the model,
585 because both pollutants are incorporated into the ice mostly at the expense of storage in soil.
586 Similarly, Stocker et al. (2007) have modelled the impact of snow and ice cover as a shield for
587 underlying soil and seawater, finding that the barrier was responsible for between 3% (for
588 α HCH) and 73% (for HCB) decrease in soil concentration against the ice-free world. The
589 seawater in the Arctic was even more influenced by the ice cover, containing only between
590 8% (for α -HCH) and 21% (for dacthal) of the hypothetical POPs amounts in the ice-free
591 world. The world with the contemporary ice cover produced higher levels of atmospheric
592 pollution (between 2 and 10 times higher, for HCB and PBDE-209, respectively).



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

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

614 Bogdal et al. (2010) pioneered in explaining englacial fate of POPs with a model and
615 achieved a relatively faithful reproduction of the storage and delay in the system, despite the
616 neglect of transport in meltwater. The reverse order (in time) of pollutant incorporation
617 into the glacial ice and its release was consistent with glaciological understanding. The

618 authors have estimated that due to climate warming, 45% of the stored POPs have been
619 released from Oberaar glacier before 2010, as opposed to 21% in the scenario with stable
620 climate. Furthermore, the changing climate has generated peak emission events beyond the
621 scale expected in the stable scenario.

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

632 Interestingly, ice flowlines explained the high concentrations in the sediment in the
633 1990s, although not the high concentrations in the 2000s. The proposed explanation for this
634 discrepancy was processes in the ablation area unaccounted for in the model. The proposed
635 delay of PCB delivery into the lake (through, for example, storage in the porous ice layer on
636 the glacier surface) was estimated for 10 years (Steinlin et al., 2016), which could be partly
637 explained by the residence time of the particulate organic matter on the glacier surface as
638 cryoconite (Kozioł et al., 2019; Takeuchi et al., 2010). Vertical meltwater percolation within
639 the glacier (not included in the model) could also relocate chemicals (Steinlin et al., 2016).
640 The model predicts a constant concentration of approximately $500 \text{ pg L}^{-1} \Sigma\text{PCBs}$ in the
641 glacial stream during the next decades, which is three orders of magnitude lower than the
642 concentration considered chronically toxic for fish; the impacts on higher levels of the trophic

643 chain need further exploration. Steinlin et al. (2016) claim also that the underestimated
644 sorption of lower-chlorinated congeners and the dynamic system of glacial lakes (Nellier et
645 al., 2015) necessitate the inclusion of biochemical processes in the model to explain the
646 accumulation of POPs in lake sediments.

647 **7. Chemical hazards in glacial melt?**

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

656 ***7.1. The quality and uniformity of the published data***

657 As the research in this field developed, the methods have been adjusted and as a result
658 the data recorded are not consistent in format across the reviewed works. We describe this
659 here as an important limitation to hemisphere-wide conclusions. A typical problem is
660 differing and non-comparable units, especially of fluxes, e.g., [$\mu\text{g}/\text{m}^2/\text{y}$], and concentrations.
661 In some cases, it was possible to calculate concentrations from fluxes according to standard
662 procedures, including e.g. snow water equivalent, ice density and age, or sediment deposition
663 rate, yet there were instances where unreported raw data hindered our efforts. Some values
664 were available on graphs alone, which decreases the accuracy of reporting. Another difficulty
665 for comparison is the common practice to administer only the sum of the concentrations of
666 compounds for such groups as HCHs or DDTs. In the case of PCBs, the problem is

667 exacerbated because the sum of PCBs may involve a different number of compounds,
668 depending on the study design. For individual PCBs, there are two main ways to present the
669 results: 1) reporting concentrations for 7 indicator PCB compounds (PCB 28, PCB 52, PCB
670 101, PCB 118, PCB 138, PCB 153, PCB 180, also named the Dutch seven); 2) grouping
671 PCBs and providing the sums of compound concentrations with the same number of chlorine
672 atoms in the molecule. PAHs experience similar problems as PCBs in reporting the sums.
673 Finally, the concentrations of individual compounds were sometimes given only in the form
674 of an average or a median, which complicated the interpretation, especially as maximum
675 concentrations are of concern for human health impacts.

676 *7.2. The consequences of the pollution trends in time*

677 The rising temperature and enhanced melt release POPs accumulated in glaciers into
678 the environment. This phenomenon has been observed in two ways: in glacial meltwater
679 (through contrasts in concentrations between snow and ice melt and between glacial and non-
680 glacial streams ; Bizzotto et al., 2009) and in sediment cores from proglacial lakes (as delayed
681 secondary concentration maxima; Bettinetti et al., 2011; Bogdal et al., 2009; Sun et al., 2018).
682 Fig. 6 shows the content of \sum DDTs, \sum HCHs, and \sum PCBs in the sediment cores, representing
683 the data for multiple sites in the European Alps and the Himalaya. The release of pollution
684 began at the end of the 1980s in the Himalayas and at the end of the 1990s in the Alps (except
685 Lake Num Co in the Himalaya, where the pollution was also liberated from the 1990s
686 onwards). Within each group of organochlorine legacy compounds, the maximum
687 concentrations from reemission occurred at a similar time. In both regions, the content of
688 OCPs and PCBs in the layers corresponding to reemissions was equal to or even higher than
689 in the layers from the peak use period. In the Alps, concentrations increased steadily until the
690 period of core collection for all POP groups. It was different in the Himalayas: the POP
691 content decreased there in the early 21st century, although it was not uniform for all sites and

692 compounds. Based on these data, POPs re-emission appears to be ongoing. However, there
693 remain important data gaps, especially profiling a wider scope of contaminant concentrations
694 and including areas such as the Arctic, the Rockies, or the rest of Asia.

695 [Figure 6 here]

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

713 PAHs ice core records are characterized by a different distribution: an underlying slow
714 increase in time (since 1970s), with overlying high extremes of short duration (some
715 overlapping across multiple locations). The slow increase can be linked to the global
716 consumption of fossil fuels and economic growth, while changes in fossil fuel type (especially



717 from coal to petroleum and natural gas) mark drops in these trends (Peters et al., 1995;
718 Vecchiato et al., 2020; Wang et al., 2008b). The short-term maxima may be linked to singular
719 events, such as volcanic eruptions. Especially, there can be found maxima in the hemispheric
720 PAHs ice core record closely coinciding with the eruptions: 1963 Surtsey (Iceland) & 1964
721 Scheveluch (Kamchatka), 1970 Hekla (Iceland), 1978 Westdhal (Alaska) & 1980 (Mt. St.
722 Helens, Washington, USA), and perhaps also the 1989 Redoubt (Alaska) and 1991 Pinatubo
723 (Philippines). The consequences of such a complex temporal pattern are: difficulties in
724 predicting the exact timing of reemission of PAHs from melting glaciers, regional differences
725 in the impacts of such a phenomenon, and its long duration due to the thicker layer being
726 affected by elevated PAHs concentrations.

727 [Figure 7 here]

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

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

752 ***7.3. Risk assessment strategies concerning the impact of glacial POPs release***

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

762 The presence of POPs in living organisms, especially in fish, and in drinking water,
763 raises the question of possible harm to human health. Unfortunately, to our knowledge, very
764 few authors have undertaken an assessment of the risk that glacial meltwater poses for human
765 health. Quadroni & Bettinetti (2017) reported that PCB levels in the fish tested sometimes
766 exceeded European Union (EU) standards regarding potential carcinogenic effects. Risk



767 assessment carried out by K. R. Miner et al. (2019) and K.R. Miner et al. (2018) in the Swiss
768 Alps and Alaska, using the Environmental Protection Agency (EPA) methodology, shows that
769 there is no risk associated with the consumption of proglacial water. However, the
770 concentrations in these waters are high enough that the consumption of fish (which
771 bioaccumulate DDTs, HCHs, and PCBs) leads to a risk of cancer. Such risk is greater for
772 children than for adults because of their lower body weight and the expected longer exposure
773 time. At the same time, in waters from another glacier Lys, in the Italian Alps, a lack of risk
774 was concluded for both fish and water consumption, in terms of their DDTs and HCHs
775 concentrations (Miner, 2018). This highlights that the assessed risk level cannot be
776 generalized at a regional scale at the moment, and warrants future research with a wider
777 spatial coverage.

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

789 The aforementioned water quality criteria indicate the risk of cancer associated with the
790 consumption of aquatic organisms and water. Using this criterion, which is based on
791 carcinogenicity risk 10^{-6} , we also scanned the collected database for potential threats. These

792 recommended concentrations of pollutants were exceeded in many cases. The contents of α -
793 HCH (Bizzotto et al., 2009; Ferrario et al., 2017; Lafrenière et al., 2006; Villa et al., 2006a),
794 HCB (Bizzotto et al., 2009), dieldrin (Lafrenière et al., 2006), and p,p'-DDT (Bizzotto et al.,
795 2009; Ferrario et al., 2017; Villa et al., 2006a) were exceeded at a maximum by one order of
796 magnitude. Moreover, PCB (Bizzotto et al., 2009; Ferrario et al., 2017; Lafrenière et al.,
797 2006; Pavlova et al., 2016) and p,p'-DDE (Bizzotto et al., 2009; Ferrario et al., 2017)
798 guideline concentrations were exceeded by almost three orders of magnitude. This fact
799 encourages the continuation of screening for POPs concentrations in glacial meltwater.
800 However, due to the fact that the highest concentrations of PCBs, HCB, and p,p'-DDE in
801 Bizzotto et al. (2009) occurred in the snow melt period (June) and the highest concentrations
802 of α -HCH, dieldrin, and PCBs were noted by Lafrenière et al. (2006) in lysimeters collecting
803 snow melt water, we vouch for including snow elution patterns in the screening study design.

804 *7.4. Recommendations for the chemical monitoring of glaciers*

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

814 POPs release from glaciers has been confirmed locally (and regionally in the European
815 Alps), and it is necessary to confirm its occurrence elsewhere, especially since risk levels may
816 differ between glaciers from the same region (Miner, 2018; Miner et al., 2018a). A screening

817 for areas already exposed to POPs release from glaciers (and the chronic toxicity risk) could
818 be based on proglacial lake bottom sediment coring (due to the efficient capture of POPs by
819 sediments). The regions especially vulnerable now and in the future may be located through
820 the interpretation of global chemical fate models, which are increasingly successful at
821 incorporating cryospheric component into the POP cycle, although their spatial distribution
822 could be further refined. Incorporating the functioning of the glacial systems in these models
823 (e.g. ELA shifts, accumulation and ablation rates, glacier geometry) is the next challenge for
824 progress in understanding glaciers as POPs storage.

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

835 **8. Conclusions**

836 The threats connected to the melting of glaciers include multiple physical effects, although
837 the chemicals released from glacial melt can also be dangerous. The increased concentrations
838 of POPs were observed both in sediment cores from proglacial lakes and in glacier-fed
839 streams (as compared to non-glacial streams). Such evidence comes mainly from the
840 European Alps and Canadian Rockies. To overcome this sampling bias, we systematically

841 collected published data on four POP class concentrations in glacial environmental media of
842 the Northern Hemisphere.

843 While the Canadian Arctic and European Alpine glaciers appear as the most polluted in
844 the hemisphere, and lower pollution concentrations occur in the Himalayas, the regional
845 concentration ranges frequently overlap, spanning <LOD to several ng/L (for some OCPs and
846 the sum of PBDEs). Only the PAHs content in glaciers exceeds that level, and it is clearly the
847 highest in Canadian Arctic at 35-660 ng/L, with intermediate levels in the Caucasus at 49-166
848 ng/L, the Himalayas at 8.9-100 ng/L or Greenland at 2-103 ng/L, and the lowest in the Alps at
849 0.5-5.2 ng/L (if locations close to high-emission hotspots are excluded). Based on the existing
850 data, the greatest threats associated with the release of POPs will take place in the Canadian
851 Arctic and the Alps, also due to the presence of thicker ice layers in the maximum emission
852 period. However, multiple regions are too poor in data for a conclusive assessment.
853 Especially, the tentative conclusion of maximum legacy POPs concentrations around 45°N
854 (Miner et al., 2017) is not maintained in light of a wider dataset. Therefore, a more
855 comprehensive way to account for the distance from POPs sources, including local hotspots,
856 and the emissions changes in time needs to be used for spatial assessments (cf. Friedman and
857 Selin, 2016; Kallenborn et al., 2012). Finally, the global pollution shifts and the remaining
858 cryosphere extent will increase the importance of monitoring both legacy and new POPs in
859 the Arctic.

860 Although the compounds we discuss are toxic, no immediate toxic effects associated with
861 their release in meltwater are to be expected. They may, however, cause harm through long-
862 term effects related to their ability to bioaccumulate and biomagnify. This is unless serious
863 synergic effects are discovered, on which there should be future research, since the
864 overlapping emission periods of various groups of POPs increase such risk. Furthermore, risk
865 assessments are still missing for the release of PAHs and PBDEs in glacial meltwater,

866 including remote locations (Himalaya, Arctic). An excess of PAHs may occur especially in
867 volcanic areas of the Arctic (Iceland, Kamchatka). The northern Canadian Arctic and the
868 Russian Arctic may be continuous recipients of current use pollutants, as they are the only
869 areas predicted to receive extra snowfall in high-end warming climate scenarios. Arctic
870 glaciers and the Himalaya are also important prospective study areas since they are yet to
871 experience their maximum melt later on in the 21st century. Hence, estimating the moment
872 when POPs stored there are released and at what concentrations would identify whether an
873 important chemical hazard to humans or ecosystems will occur.

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

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890

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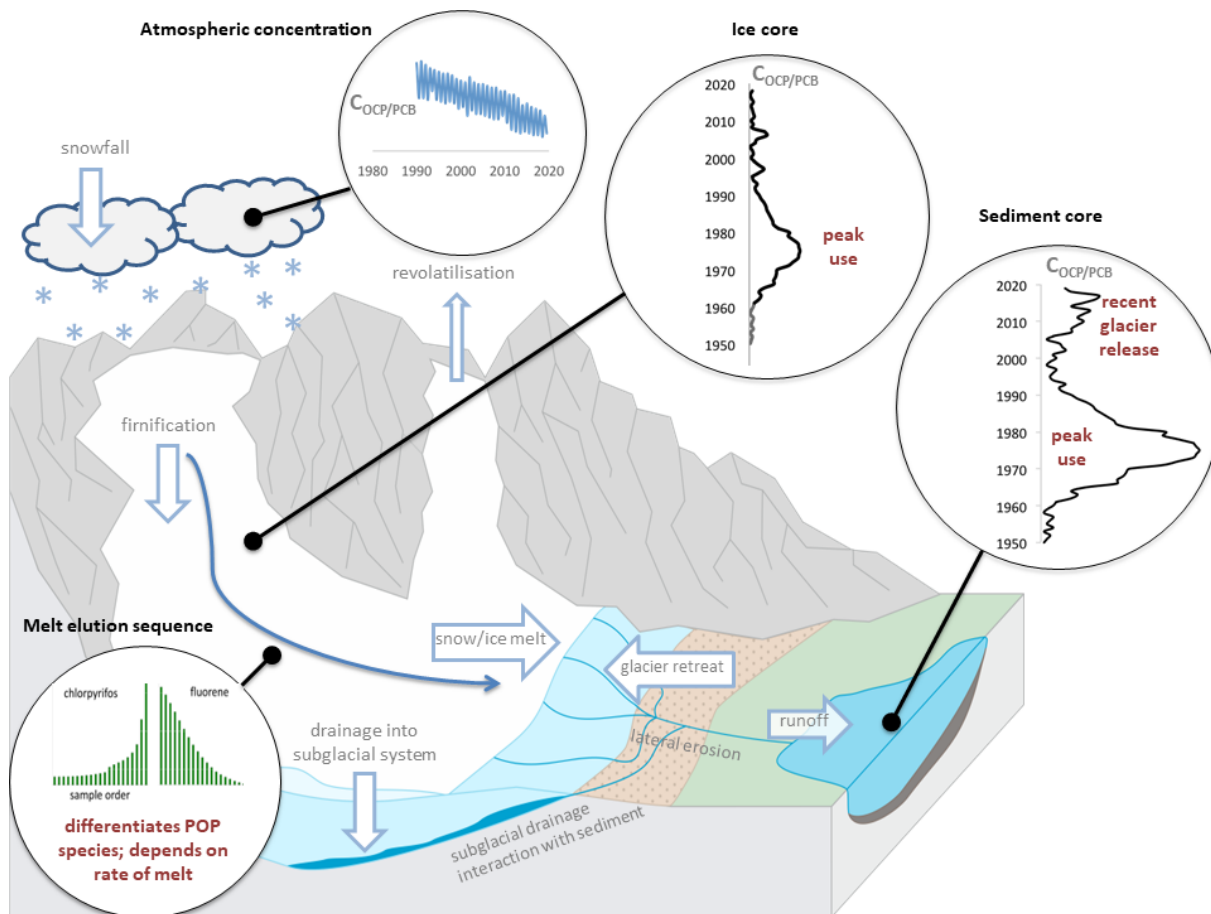


Figure 1. A diagram of the glacial system with the environmental media potentially storing pollutants (with sample pollutant concentration (C_{POP} , $C_{OCP/PCB}$) graphs).

POPs partitioning between gaseous, dissolved and adsorbed (between phases of water and attachment to impurities)

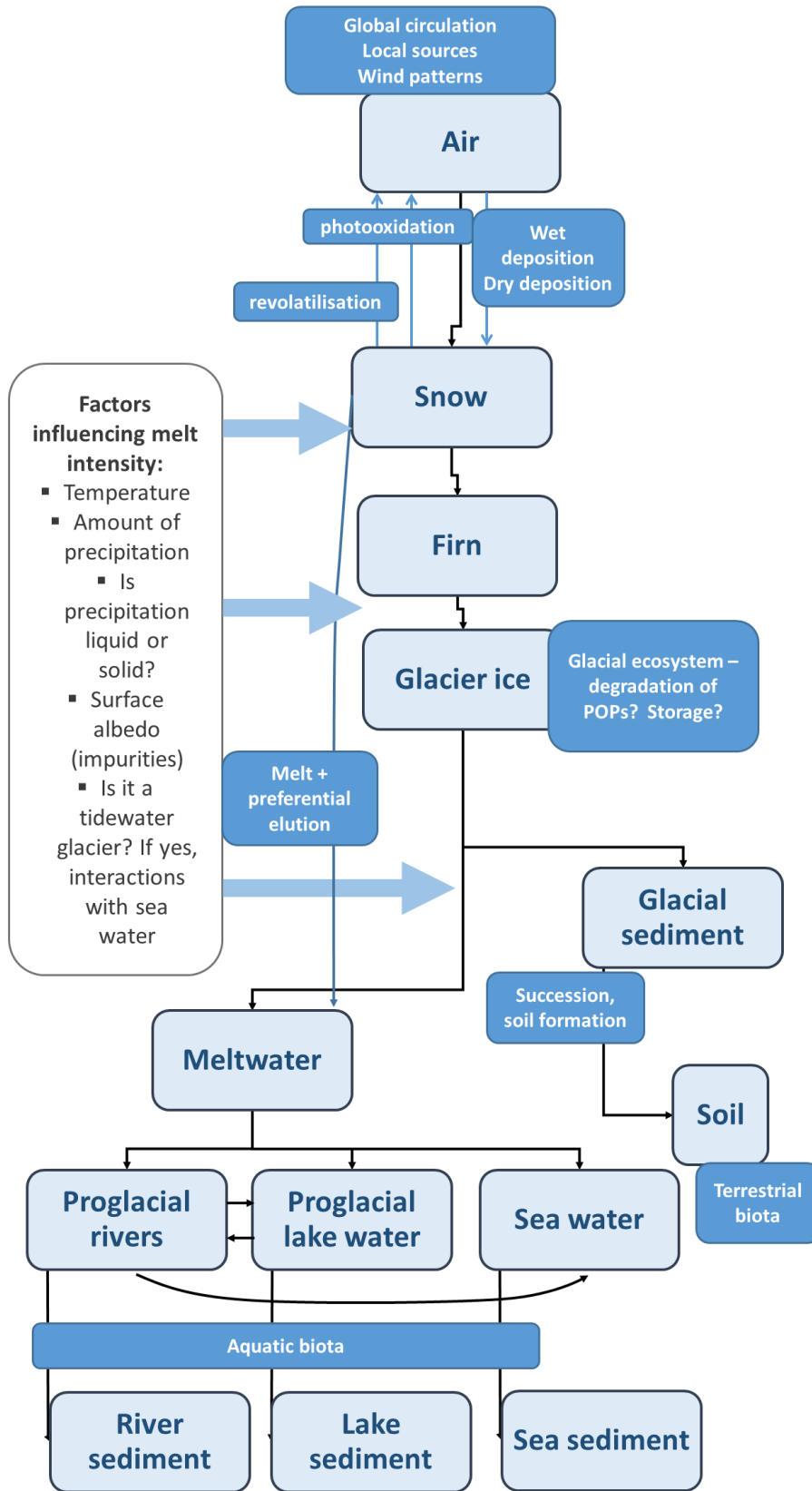


Figure 2. POPs pathways through the glacial system, including the factors influencing their transport efficiency and storage time.

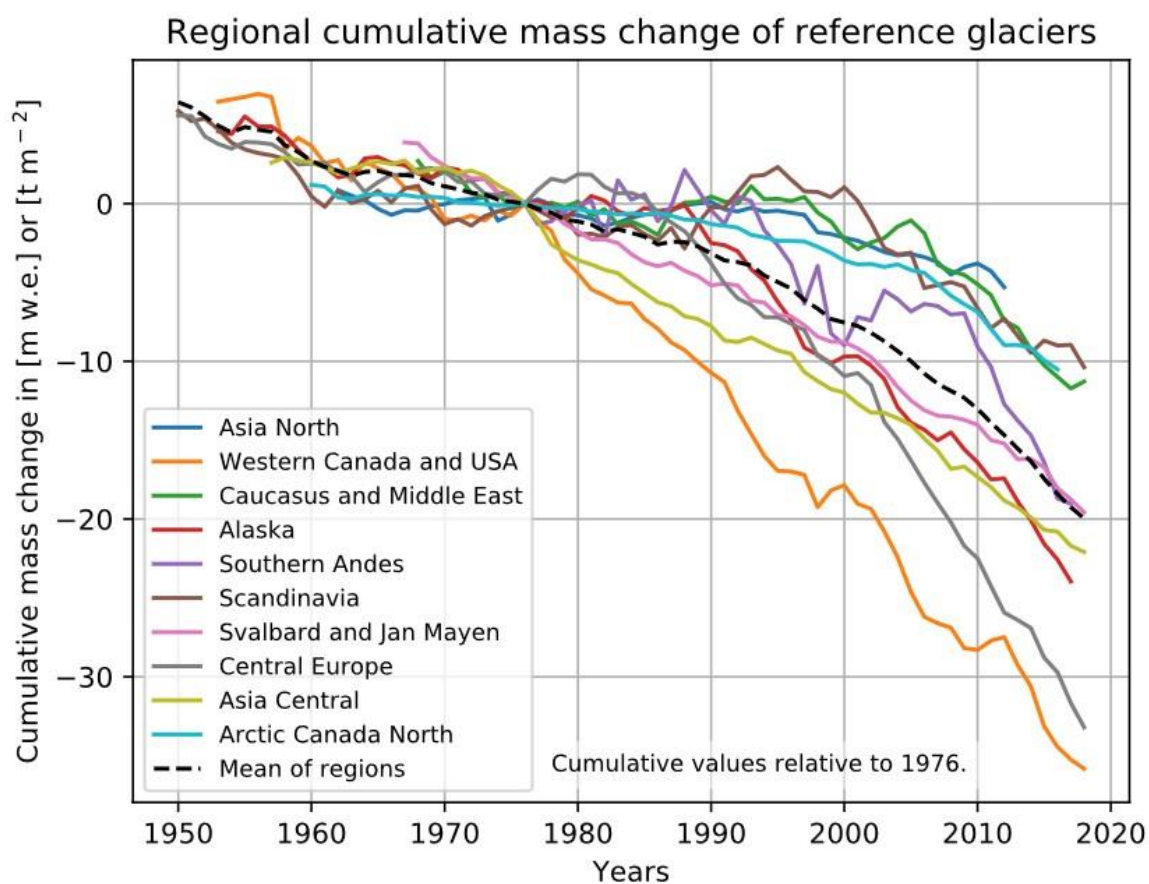


Figure 3. Cumulative mass balance change (since 1976) in glaciers around the world, divided by region. Source: (WGMS, 2020). *Reprinted with permission.*

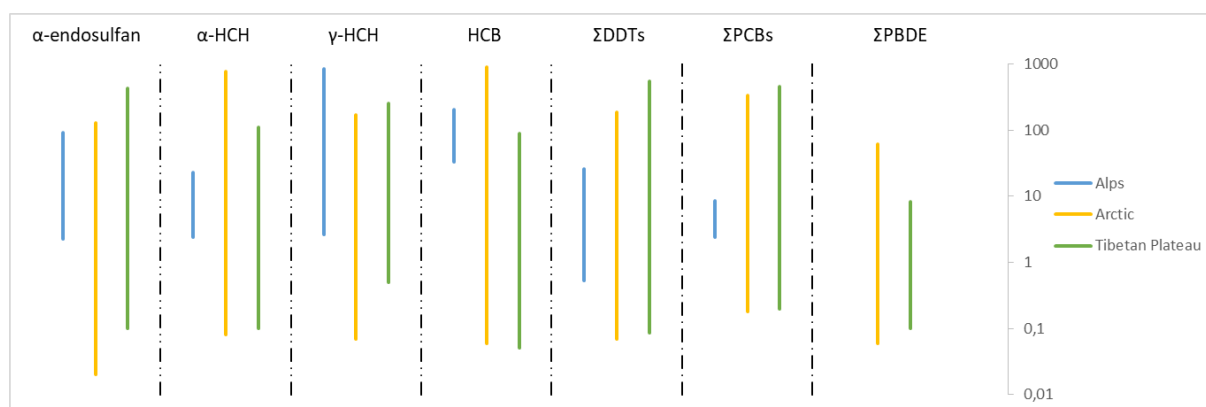
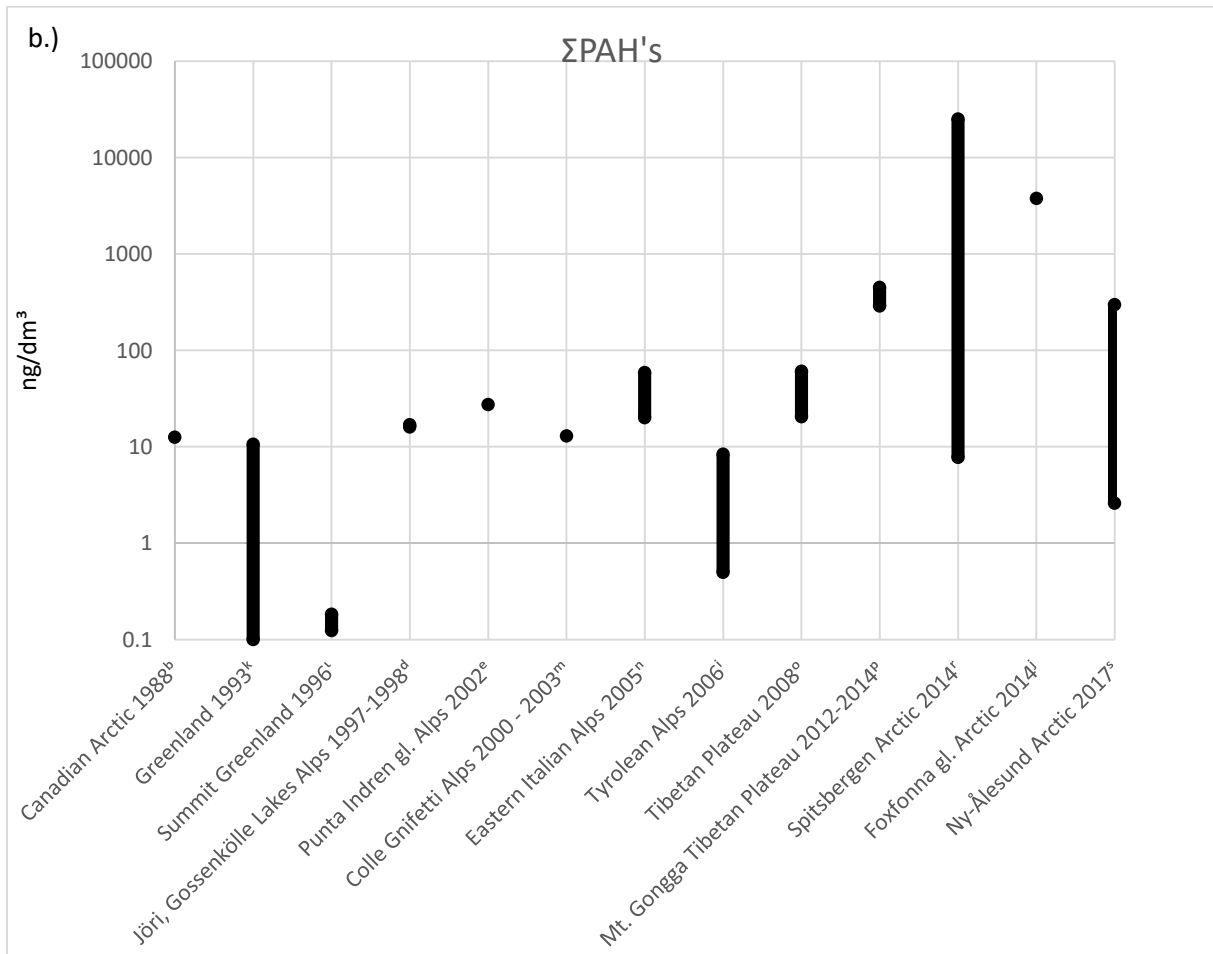
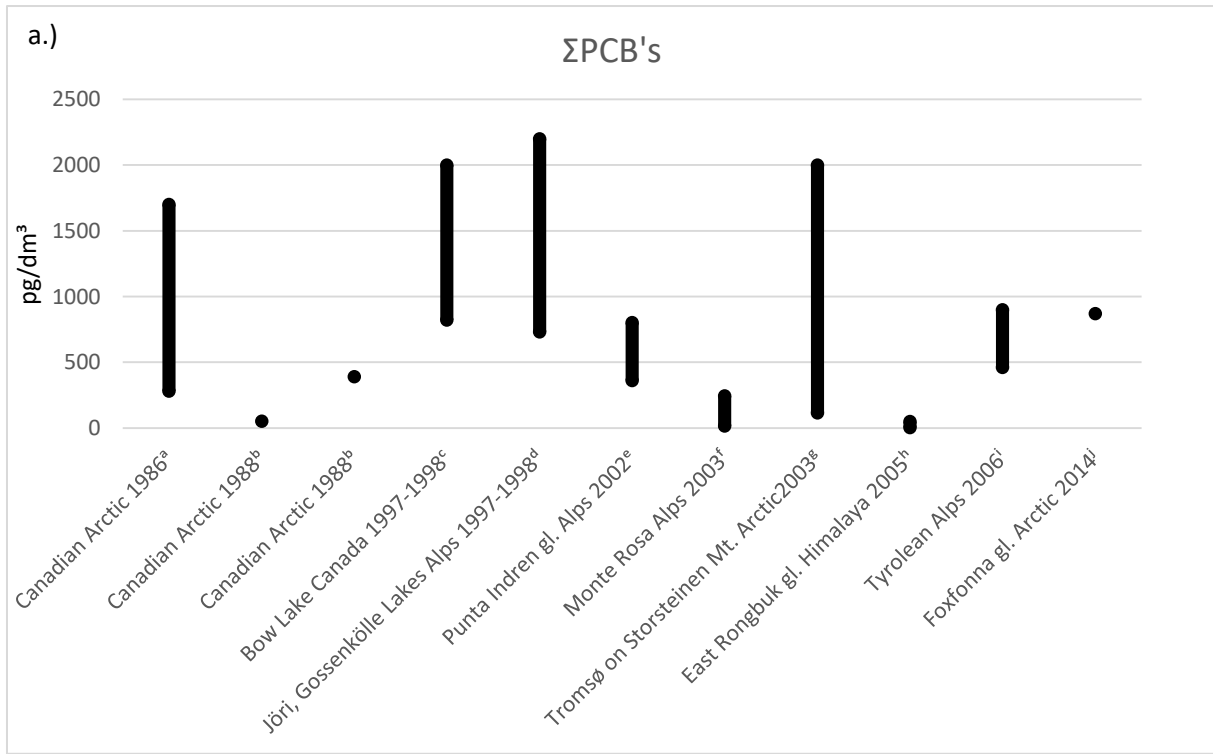


Figure 4. Comparison of the concentration ranges [pg/m³] of atmospheric POPs in the Alps, Arctic and Tibetan Plateau. Data and references used in this figure can be found in Table S2-3.



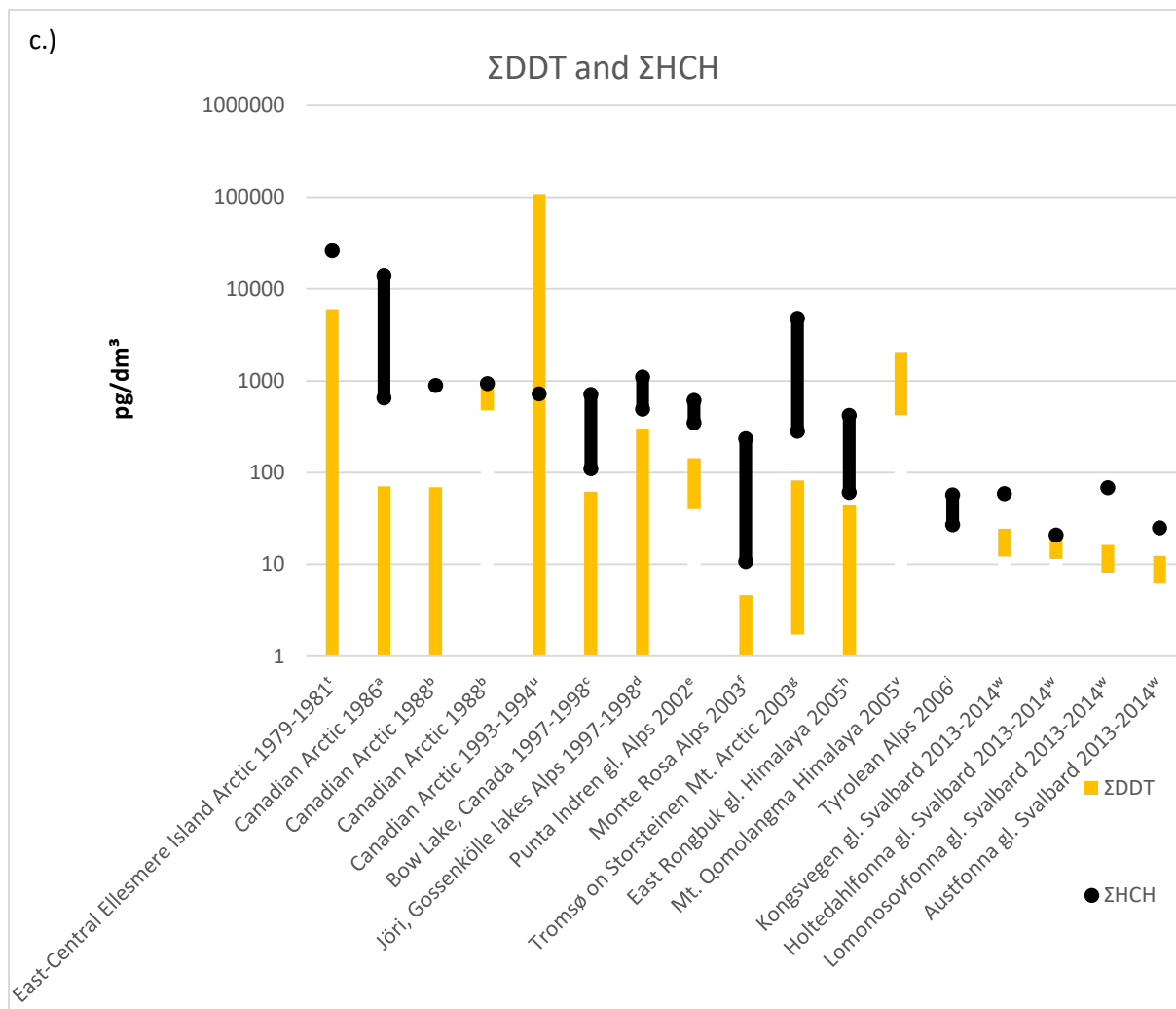
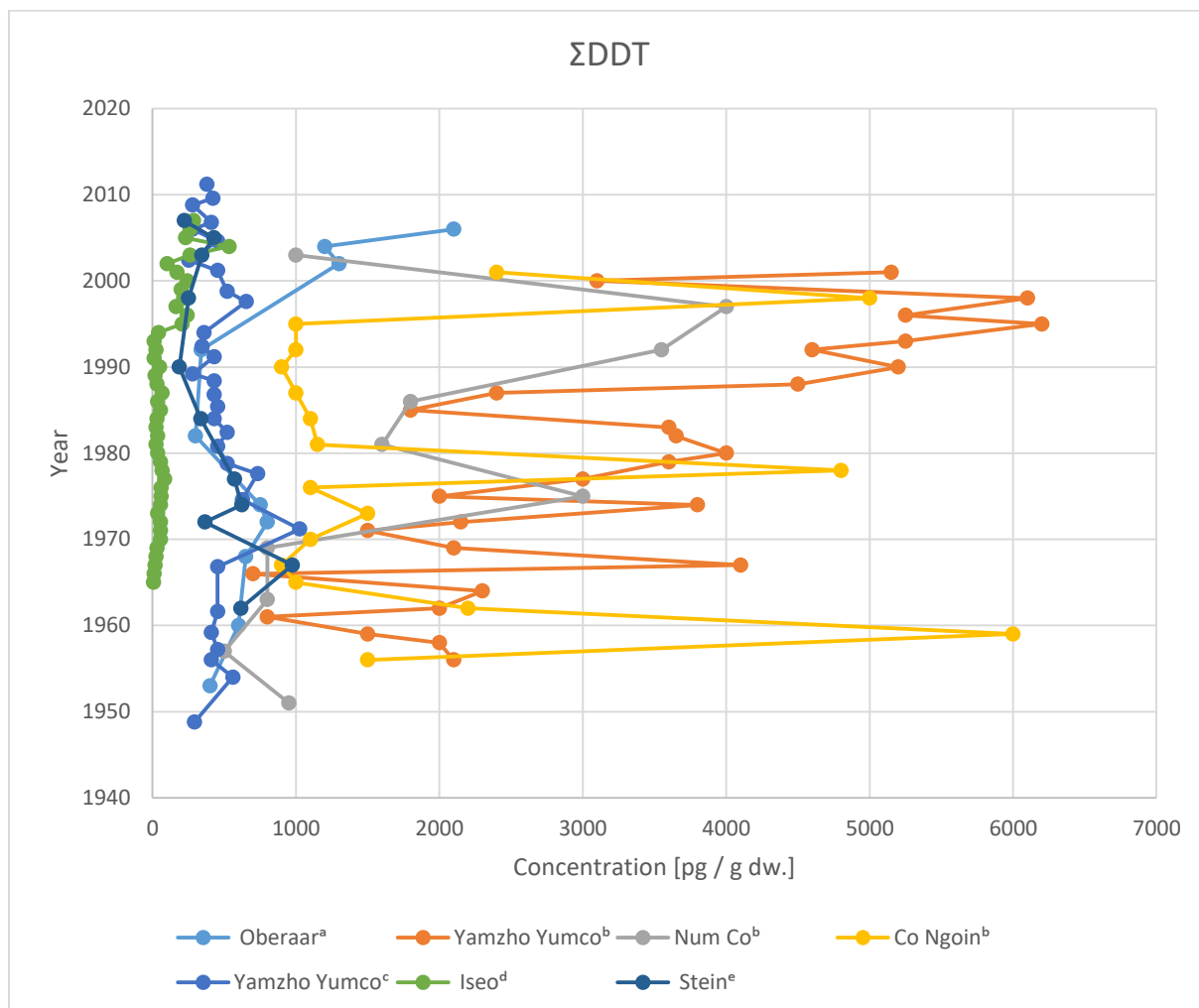
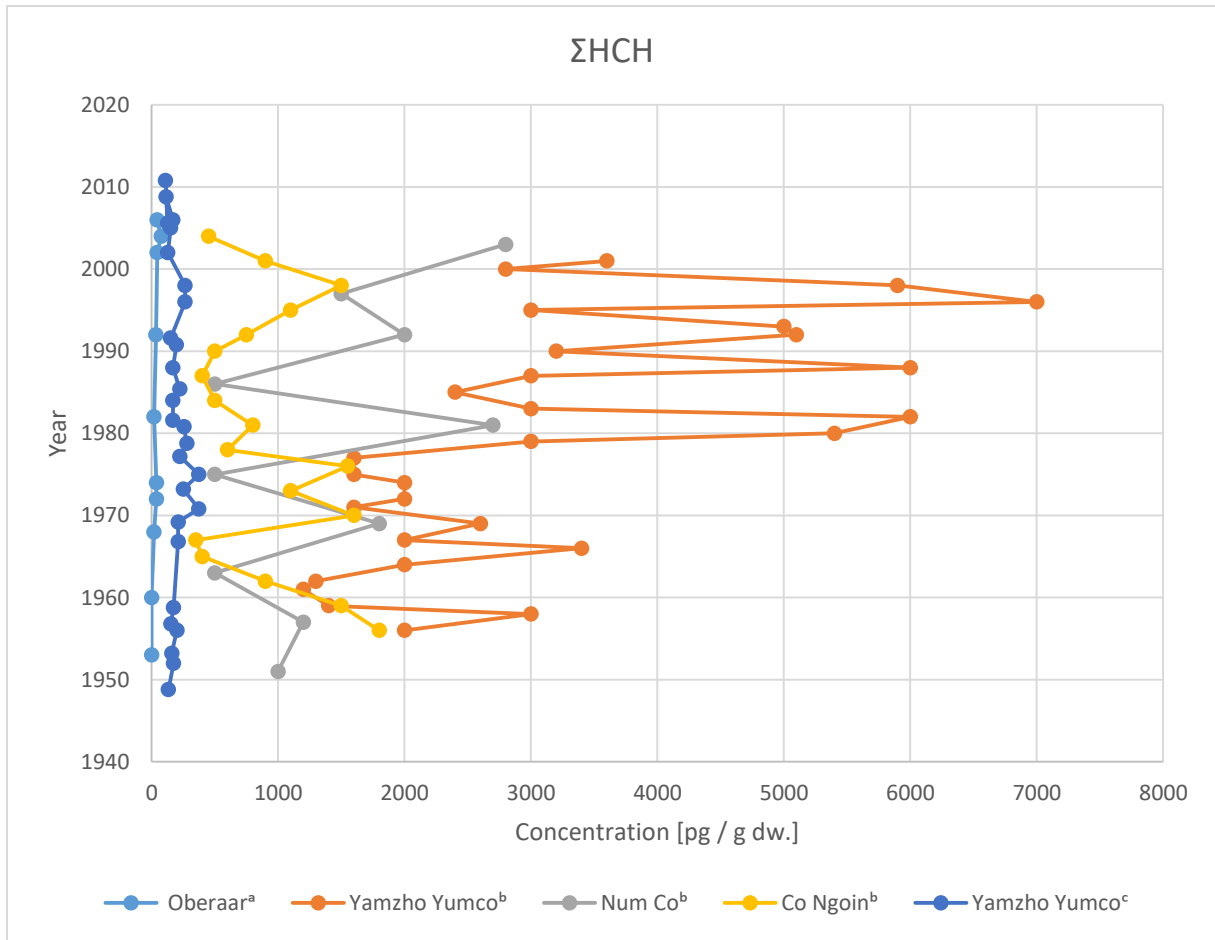


Figure 5. Range of a.) Σ PCBs [pg dm⁻³ of melted snow water], b.) Σ PAHs [ng dm⁻³ of melted snow water] and c.) Σ HCHs and Σ DDTs concentration in [pg dm⁻³ of melted snow water] concentration in snow in various glaciated regions worldwide. a - (Dennis J Gregor & Gummer, 1989), b - (Welch et al., 1991), 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 - (Masclat 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).





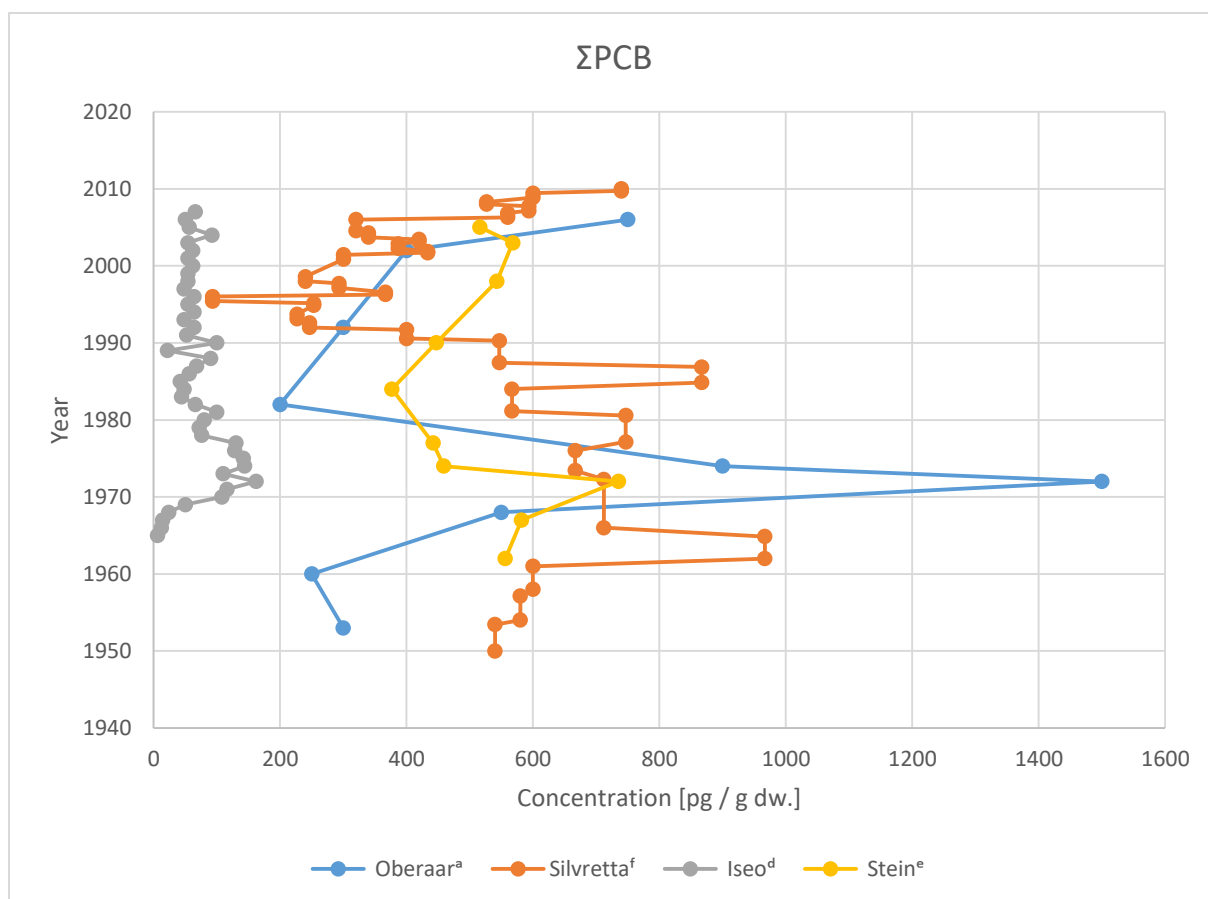
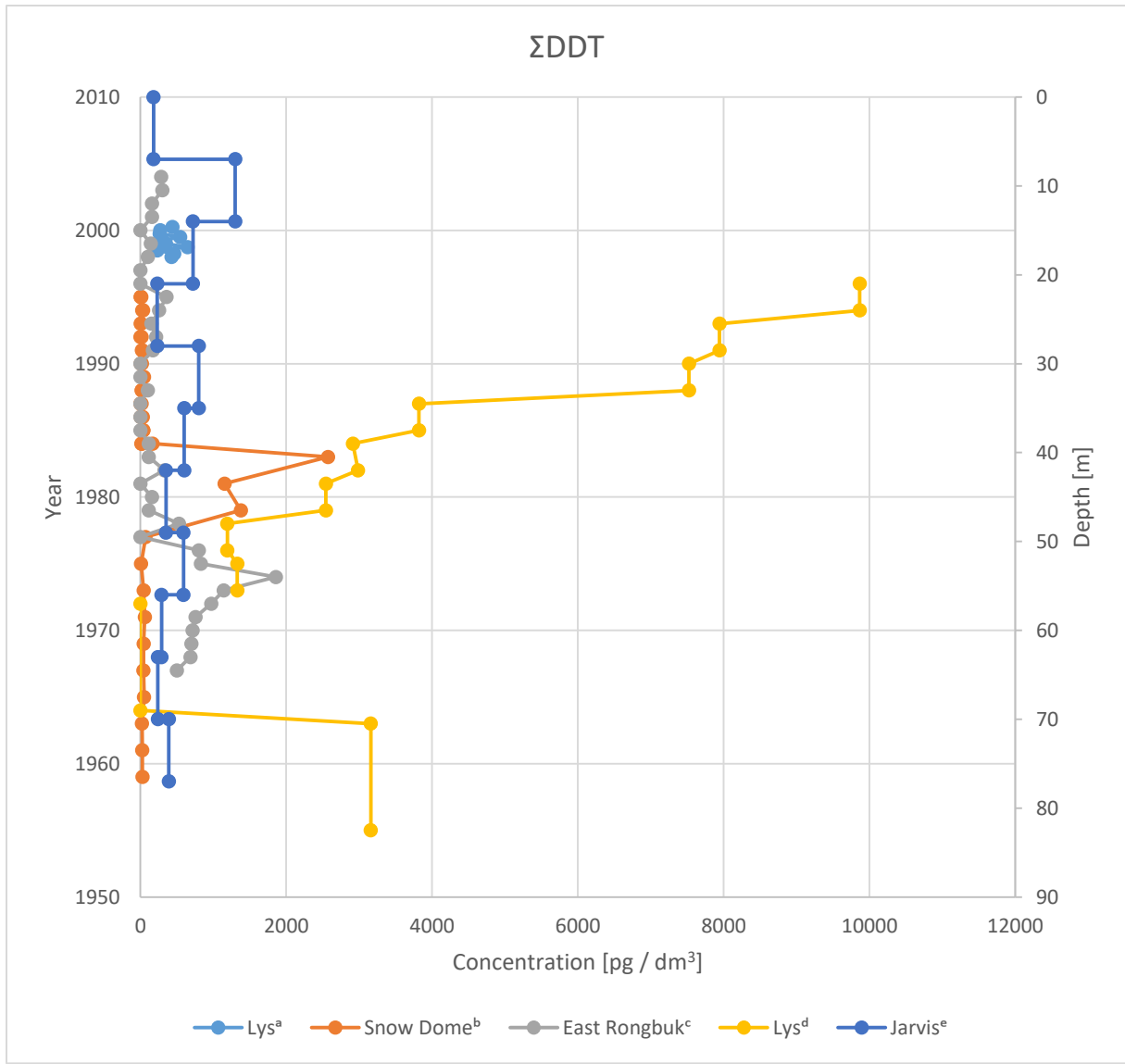
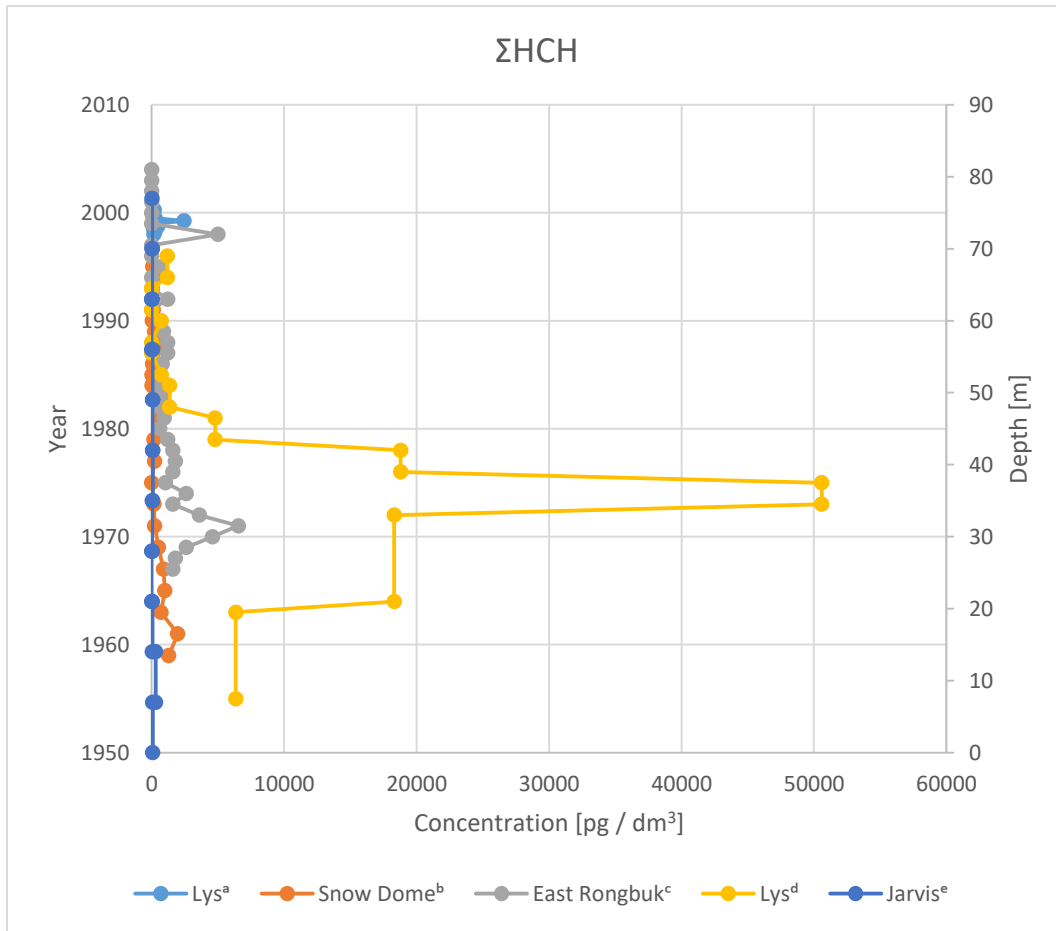
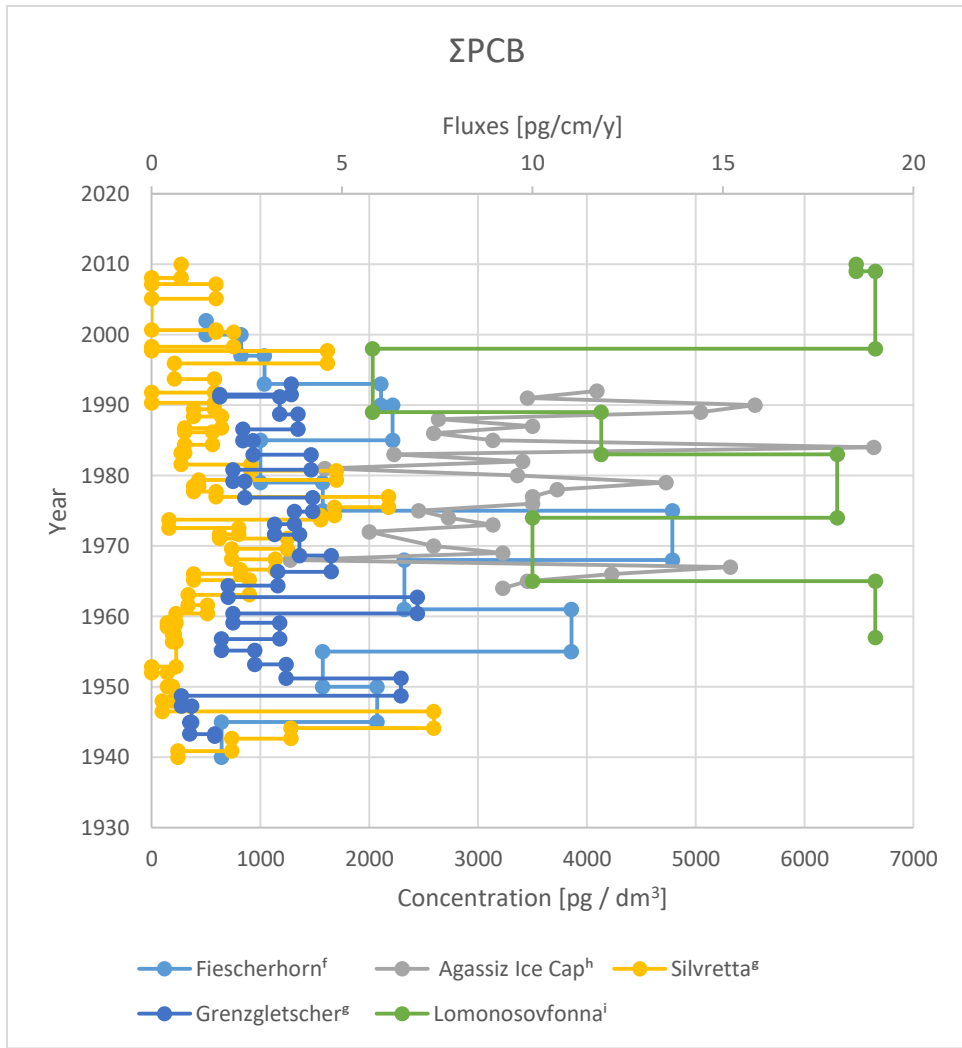


Figure 6. Concentrations of Σ DDT, Σ HCHs, Σ PCBs in the sediment cores from proglacial lakes in the European Alps and Himalaya. a – (Bogdal et al., 2009), b – (Cheng et al., 2014), c – (Sun et al., 2018), d – (Bettinetti et al., 2011), e – (Schmid et al., 2011), f – (Pavlova et al., 2016), e – (Schmid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







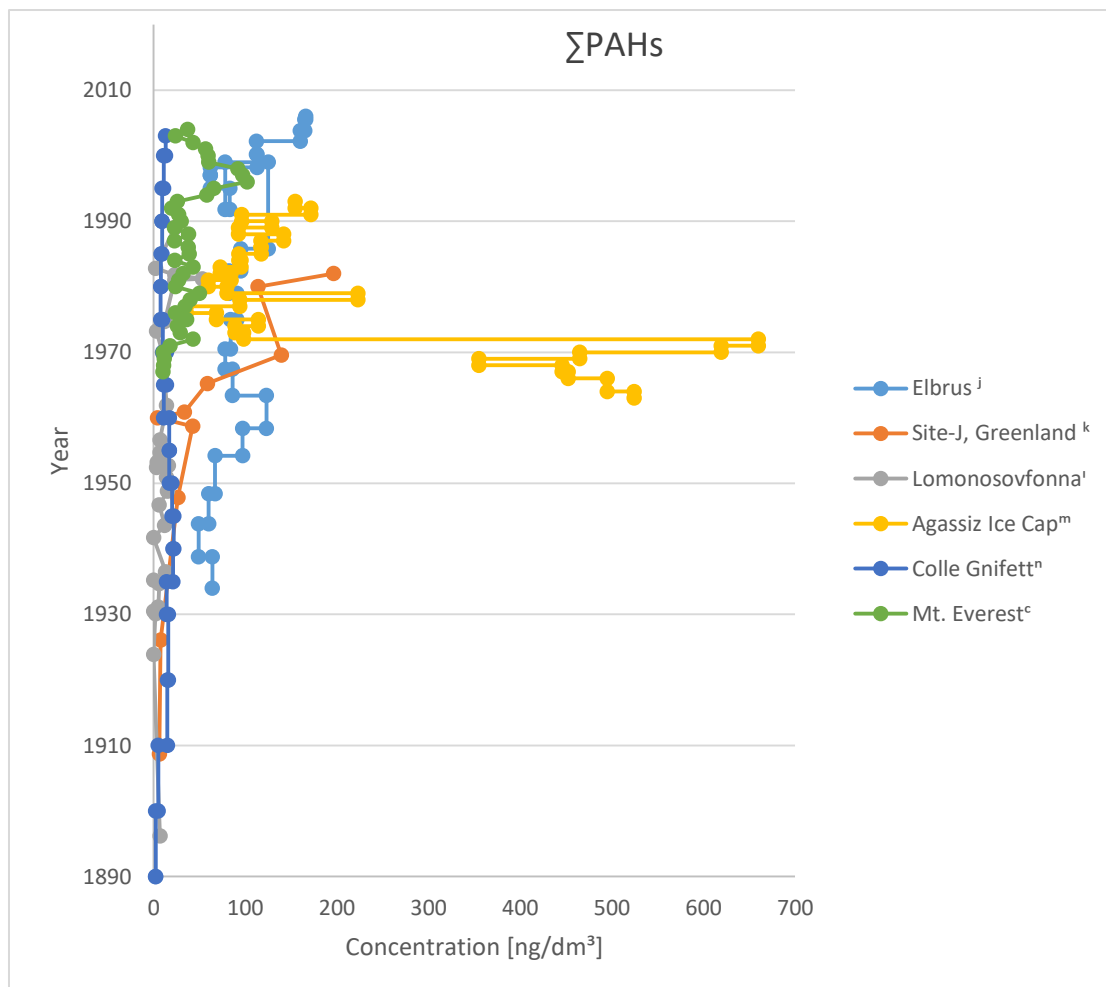


Figure 7. Concentration of Σ DDTs (please note: the age of layers was not specified for Jarvis glacier, hence the concentrations are plotted using depth), Σ HCHs (Jarvis glacier concentrations are plotted by depth instead of age), Σ PCBs (please note that the contents of the Σ PCBs for Lomonosovfonna should be read from the top, secondary axis (as fluxes)), Σ PAHs 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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