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10 **A High-Arctic flow-through lake system hydrochemical changes: Revvatnet,**  
11 **southwestern Svalbard (years 2010-2018)**

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19

20 **Abstract:** Lake ecosystems are strongly coupled to features of their surrounding landscapes  
21 such as geomorphology, lithology, vegetation and hydrological characteristics. In the 2010-  
22 2018 summer seasons, we investigated an Arctic flow-through lake system Revvatnet, located  
23 in the vicinity of the coastal zone of Hornsund fjord in Svalbard, characterising its  
24 hydrological properties and the chemical composition of its waters. The lake system  
25 comprises of a small upper lake and a large lower one, the latter cone-shaped, with -29.1 m  
26 maximum depth. With near-neutral pH (full range 6.5 – 8.4) and low EC (7 to 147  $\mu\text{S cm}^{-1}$ ),  
27 the lake has rather similar characteristics to many Arctic lakes. Metal and metalloid  
28 concentrations were either similar across the lake system or increased downstream (except Zn,  
29 which has important ore-bearing veins in the upper part of the catchment), which is consistent  
30 with the likely slow dissolution of suspended particles within the lakes. The  $\Sigma\text{PAHs}$   
31 concentrations ranged from <MDL to 2151  $\text{ng L}^{-1}$ , and according to the indicator PAHs  
32 concentration ratios, they originated from a mixture of combustion processes (they were not  
33 petrogenic). Principal component analysis showed that seasonal variability was the most  
34 characteristic feature of the chemical composition of these waters, although there appear to be

35 consistent changes with time (sampling year) as well. Future research should explore the  
36 occurrence of high maxima in the concentrations of priority pollutants, such as PAHs, metals  
37 and metalloids (e.g. As).

38

39 **Keywords:** Arctic; flow-through lake; bathymetry; freshwater; chemical pollution

40

## 41 **1. Introduction**

42 A characteristic and often dominant feature of polar landscapes is the great diversity and  
43 abundance of standing surface waters (Kling, 2009). Many lakes owe their origin to glacial  
44 action that shaped their lake basin and surrounding landscape, and that distributed glaciogenic  
45 deposits of different sources and mineral composition across their catchments, a fact  
46 referenced by other studies, e.g. Marszałek and Górnjak (2017). In the postglacial landscape,  
47 lake density is more than four times higher than in terrain which was not glaciated (Smith et  
48 al., 2007). Furthermore, lake densities and area fractions are on average approximately 100 –  
49 170 % greater in permafrost-influenced terrain (compared with permafrost-free) (Vincent and  
50 Laybourn-Parry, 2008).

51 High-latitude lakes are likely to show ongoing responses to present climate instability that  
52 leads to deglaciation, variations in chemical composition of freshwater, and, for lakes located  
53 close to coastal waters, changes in their linkages with the sea. All of these processes are major  
54 controls on the structure and functioning of aquatic ecosystems (Van Hove et al., 2006;  
55 Vincent and Laybourn-Parry, 2008). Geological variety of the polar regions, including the  
56 differences in substrate lithology, may also affect the extent of rock weathering and the  
57 chemical composition of soil water that finally discharges into polar lakes (Hamilton et al.,  
58 2001; Szumińska et al., 2018; Vincent and Laybourn-Parry, 2008). In brief, Arctic lake



59 ecosystems are strongly coupled to the features of their surrounding landscapes, such as  
60 hydrological characteristics, geomorphology, lithology and vegetation, and due to this, there  
61 are several significant differences in lake characteristics in various parts of the Arctic (Wetzel,  
62 2001).

63 High solar radiation levels, reaching high latitudes in spring, result in rapid snowmelt. Runoff  
64 in late spring typically comprises 80 – 90 % of the annual total in the Arctic, while it lasts  
65 only a few weeks (Prowse and Ommanney, 1990). Summer sources of freshwater include  
66 perennial and durable snow patches, rain, glaciers, melting of the active permafrost layer and  
67 some cases of groundwater discharge (Vincent and Laybourn-Parry, 2008). Groundwater  
68 levels and distribution within polar regions are greatly influenced by bedrock geology,  
69 permafrost layers and soil thickness (Prowse and Ommanney, 1990).

70 Freshwater is crucial to the unique and fragile ecosystems of the Arctic. Climate change  
71 enhances the moisture transport from lower latitudes towards the pole (Mueller et al., 2009).  
72 This is contributing to increases in precipitation in the Arctic, falling either as rain or snowfall  
73 and transporting at the same time a variety of persistent organic pollutants (POPs),  
74 characterised by durability and resistance to degradation. The residence time of those  
75 pollutants is long enough for them to be transported thousands of kilometres by air and ocean  
76 water (Halsall et al., 2001). Consequently, the compounds that have not been produced for the  
77 past few decades still appear in the environment (including polar regions). The quantities of  
78 them may impact negatively on the functioning of ecosystems, animal and human health  
79 (Kozak et al., 2013). In many parts of the Arctic, the proportion of precipitation that falls as  
80 rain, as opposed to snow, has increased, and the period of snow-cover has become shorter  
81 (Kling, 2009; Olichwer et al., 2013). These processes have a significant impact on the  
82 hydrological changes of the entire Arctic lake catchments and they are expected to continue,  
83 although to variable degrees between different parts of the Arctic and over different seasons.

84 Among polar regions, the Svalbard archipelago is distinguished by its location as the gateway  
85 to the Arctic. Relatively close location of Svalbard archipelago to Europe makes this sensitive  
86 region particularly exposed to the influence of pollutants (Kozak et al., 2016). Additionally,  
87 the landscape of the Revvatnet lake area with its prevalence of mountains, favours the  
88 accumulation of pollutants transported by air masses from Europe and Asia. Therefore, it has  
89 been chosen for comprehensive studies on the pollutants deposition (e.g., Laing et al., 2014;  
90 Wojtuń et al., 2013). Worldwide, countries are striving to reduce emissions of potentially  
91 toxic pollutants to reduce their levels in all environmental media. Therefore, studying the  
92 pollution in polar waters is a timely and important research task.

93 Since the Revvatnet is a flow-through lake, near the river estuary in the Hornsund fjord, it can  
94 be expected that some of its pollution will be transported to the coastal zone, causing adverse  
95 effects. Many contaminants are particle-bound, thus sedimentation and resuspension  
96 processes will determine whether they reach the coastal zone and the sea or if they are  
97 deposited along the way. When a river flows slowly, particles usually settle, and bottom  
98 sediments can become enriched in persistent organic pollutants (e.g. the PAHs described  
99 here). However, the river bottom is usually only a temporary trap, since turbulent flow would  
100 lead to the resuspension of particles (CliC/AMAP/IASC, 2016). The objective of the present  
101 study was to characterise the hydrological properties of the High-Arctic flow-through  
102 Revvatnet lake in Svalbard and to evaluate its chemical composition, especially of  
103 contaminants, in an eight-year period (2010-2018), taking into account geomorphology . The  
104 latter, as well as pollutant accumulation, both have the potential to affect lake evolution  
105 through a variety of processes, especially in the polar regions which are sensitive to small  
106 alterations in air temperatures and the chemical composition of water (Kozak et al., 2016;  
107 Szumińska et al., 2018).

108



## 109 **2. Materials and Methods**

### 110 2.1. Study area

111 Revvatnet is located in Wedel-Jarlsberg Land, in southwestern Spitsbergen (15°36'E,  
112 77°02'N), in the vicinity of the Polish Polar Station in Hornsund. The studied lake is situated  
113 at 40 m a.s.l. within the glacial Revdalen catchment. The Revvatnet lake is fed by atmospheric  
114 precipitation, snow melt water streams and the Revelva river with its tributaries. Revvatnet is  
115 covered with ice for about 8-9 months a year. It is usually free from ice cover from mid-June  
116 to the end of September. However, the lake does not freeze to the bottom.

117 The upper part of the Revdalen catchment is characterised by the streams originating from the  
118 slopes of Eimfjellet and Skålfjellet. The catchment is characterised by many tributaries  
119 located on its left side, of which the proglacial Ariebekken is the largest (Kosek et al., 2019a).  
120 The Revvatnet, as an example of a High-Arctic flow-through lake system, has been chosen for  
121 a study on pollution transfer through such a system. For that purpose, both lake bathymetry  
122 and water chemistry have been investigated. Sampling locations for chemical measurements  
123 have been selected at the left shore of the lake, mainly to determine inputs from tributaries on  
124 that side.

### 125 2.2. Bathymetric measurements

126 Bathymetric measurements were collected in August 2010 using a Lowrance LCX-17MT  
127 echo sounder system with a teleCover-Lok telescopic handle mounted on the transom of the  
128 rubber boat, recording the path of the boat movement with an integrated GPS receiver (LGC-  
129 2000). Bathymetric measurements were made according to the methodology proposed by  
130 Lange (1993) and presented in the guide for hydrographic fieldwork (Gutry-Korycka and  
131 Werner-Więckowska, 1996). The development of the bathymetric plan of the lake required a  
132 series of cross-sections and longitudinal sections, and then the transfer of information about

133 the distribution of depth in these profiles to the digital map of the lake. The total length of all  
134 bathymetric profiles was over 20 km (2700 measuring points), which allowed to draw up a  
135 bathymetric plan as well as a three-dimensional model of the lake basin. ArcGIS 10.6.1  
136 (ESRI) and Surfer 18 softwares (Golden Software) were used to make the bathymetric plan  
137 and the lake model.

### 138 2.3. Freshwater sampling

139 Water samples were collected from the Revvatnet lake in the summer periods between 2010  
140 and 2018 (all samples were collected from the same sampling points, Figure 1). The timing of  
141 the sampling campaigns was adjusted to the seasonality of the atmospheric conditions in  
142 Hornsund, yet logistical constraints prevented us from repeating the sampling every year, and  
143 sometimes the month of sampling shifted. The summer is considered to be a period of first  
144 snow, then glacier ice and permafrost melting, and through these processes pollutants enter  
145 the studied lake. Moreover, there have been significant trends noted in atmospheric  
146 precipitation sums across the summer period, which may modify the delivery of pollutants  
147 through wet deposition: a decreasing trend in June, and a rising one in July and August  
148 (Wawrzyniak and Osuch, 2020). Finally, as summer biomass burning events tend to have a  
149 stronger impact on the atmospheric particulate matter in Svalbard (Zielinski et al., 2020),  
150 there is a risk of increasing polycyclic aromatic hydrocarbons (PAHs) delivery in that period.

151 **Figure 1.** Location of the sampling points Rev 1-5. For a colour figure, please refer to the  
152 online version of the article

153 Samples were collected manually from the Revvatnet lake at a distance of 1.5 m from the  
154 shore with no headspace into air-tight, chemically clean bottles (the purity of the procedure  
155 has been verified by daily blank sample collection). Pre-cleaning procedure for the bottles  
156 included week-long soaking with Milli-Q deionised water and removing the water from the

157 sampling containers several times. The water was sampled from a depth between 20 and 50  
158 cm below water level.

#### 159 2.4. Chemical analyses

160 Milli-Q deionised water (18 M $\Omega$ ) was used during the determination of the various target  
161 analyte groups to determine instrumental background (by inserting it in the analysis queue  
162 once every six samples) and to prepare sample container blanks. The concentrations of  
163 chemical compounds in the collected samples were determined by: 1) for metals and  
164 metalloids: Inductively Coupled Plasma – Mass Spectrometry (the element concentration CVs  
165 of the obtained triplicate results ranged from 0.5 to 1.5%); 2) for total organic carbon (TOC):  
166 TOC-VCSH/CSN Analyser (Shimadzu, Japan) with NDIR detector; 3) for formaldehyde and  
167 the sum of phenols: a spectrophotometer, and 4) for PAHs: a gas chromatograph coupled to a  
168 single quadrupole mass spectrometer. All the applied analytical procedures have been  
169 validated against certified reference materials (CRMs) concordant with ISO Guide 34:2009  
170 and ISO/IEC 17025:2005, and the data obtained here were subject to strict QC procedures.  
171 Prior to pH measurements, a three-point electrode calibration was performed with temperature  
172 compensation, using MERCK Millipore Certipur® buffer solutions of pH 4.00, 7.00 and 9.00  
173 (at 25°C). In the analysis of metal and metalloid concentrations, we applied the Standard  
174 Reference Material (RM) NIST 1643e Trace Elements in Water, and RM Enviro MAT ES-L-  
175 2CRM, ES-H-2 CRM SCP SCIENCE. The calibration of the apparatus was based on RMs by  
176 Inorganic ventures ANALITYK: CCS-4, CCS-6, CCS-1, IV-ICPMS-71A. Potassium  
177 hydrogen phthalate by NacalaiTesque (Japan) was used for the calibration of the TOC  
178 Analyser. Each sample was analysed in triplicate. Sensitivity tests were performed by  
179 injecting standard analyte mixtures within the measured concentration range, and linear  
180 calibration curves of the peak area against concentration showed correlation coefficients ( $R^2$ )  
181 in the range of 0.898–0.999 for all standards. Detailed technical specifications of the applied



182 determination techniques, as well as the validation parameters of the analytical procedures,  
183 are summarised in Table S1 (Supplementary information).

## 184 2.5. Statistical analyses

185 The significance of temporal trends and inter-group differences was checked in STATISTICA  
186 13 (TIBCO Software Inc.), and due to the lack of normal distribution for most of the data, we  
187 applied non-parametric statistics, such as Kruskal-Wallis ANOVA. The same software was  
188 used for exploring data distributions and the drawing of box-and-whisker plots.

189 Principal Component Analysis (PCA) is a multivariate statistical analysis that allows  
190 revealing internal relations in the data set. PCA finds linear combinations of the original  
191 variables, referred to as principal components, which provide better descriptors of the data  
192 pattern than the original measurements and account for most of the dataset variation. Principal  
193 component analysis (PCA) has been conducted in R software (version 3.6.2.), using the  
194 *FactoMineR* package. Missing data were omitted by case. Prior to analysis, values <MDL  
195 have been replaced with  $\frac{1}{2}$  of the MDL value, subsequently all variables except pH (which  
196 already is a logarithm) were log-transformed to bring their distribution closer to normal.  
197 Departures from normality were the most pronounced where there were several <MDL  
198 values. Due to the exploratory character of the performed PCA, we accepted minor departures  
199 from the normal distribution without further data transformation. All variables were scaled  
200 (i.e. z-transformed) as part of the PCA calculation procedure (by using the parameter  
201 `scale.unit=TRUE`). R was also used to calculate the Wilcoxon signed-rank test results (with  
202 the *wilcox.test* function) for the matched samples (i.e. from two sites in the same lake system,  
203 which are connected by water flow).

## 204 205 **3. Results**



### 206 3.1. Hydrological characteristics of the lake (morphometry)

207 The Revvatnet lake, with a surface area of  $0.9 \text{ km}^2$ , has an elongated shape – its axis runs  
208 NW-SE. Its maximum length ( $l$ ) is 2.40 km, and the lake elongation index ( $\lambda$ ) equals 6.4. The  
209 mean width ( $B_{av}$ ) of the lake is 0.38 km, while the maximum width ( $B_{max}$ ) reaches 0.60 km.  
210 The lake shoreline is moderately developed, with a shoreline development index ( $K$ ) of 1.6  
211 and the shoreline length ( $l$ ) of 5.37 km. The mean lake depth was approximately 9.0 m, while  
212 the maximum depth, recorded during bathymetric measurements, reached 29.1 m (Figure 2).  
213 The volume of the lake ( $V_0$ ) was calculated at  $0.0086 \text{ km}^3$ . The shape of the lake resembles a  
214 cone, which can be estimated from its relative depth ( $h_w$ ) of 30.6% and the lake depth  
215 indicator ( $W_g$ ) of 0.33. This morphometric feature is a characteristic of glacial lakes (Figure  
216 S1, Supplementary Information).

217 For the context of geomorphological situation of the lake, we refer the reader to the  
218 geomorphological map (Jania et al., 1984). In brief, the river valley form, within which the  
219 lake is located, is surrounded by plains of raised marine terraces (in the upper part of the  
220 catchment only by flat ground moraine plains to the east of the lake). Frost-wedge polygons  
221 occur in the part of the catchment surrounding and below the lower part of the lake.

222 **Figure 2.** Bathymetry of the Revvatnet lake. For a colour figure, please refer to the online  
223 version of the article

### 224 3.2. Chemical composition of freshwater samples collected in summer periods between 2010 225 and 2018

226 The chemistry of polar freshwater reservoirs is shaped by the geological substratum, as well  
227 as by atmospheric deposition, marine aerosols, chemical weathering, biological processes,  
228 glacial and periglacial activity. Depending on the geological structures and soil cover on the  
229 raised marine terraces and in the surrounding areas, the waters that reach tundra lakes

230 transport varying proportions of specific chemicals (Mazurek et al., 2012). The results  
231 obtained for the Revvatnet samples from the summer periods of 2010 to 2018 are shown in  
232 Table S3 (Supplementary information).

### 233 3.2.1. pH and electrical conductivity (EC) measurements

234 Basic physico-chemical parameters, such as pH can vary greatly in aquatic ecosystems (Bååth  
235 and Kritzberg, 2015). Lake, river and stream waters vary in pH values from below 4 to above  
236 9, even within small geographical areas. In highly productive lakes, pH at the surface may be  
237 2 units higher than in bottom waters. The variation of the values is driven by vertical  
238 differences in photosynthesis, respiration, and redox conditions (Wetzel, 2001). pH can also  
239 fluctuate rapidly. For example, during snow melt and rain storms, pH values in streams can  
240 decrease several units, sometimes within a few hours (Lawrence, 2002). On the other hand,  
241 sunny days can result in high photosynthetic activity, leading to an increase in water pH.  
242 Accordingly, changes of 2-3 pH units may be found in highly productive aquatic  
243 environments (Tank et al., 2009). In our study, the pH values in the samples collected over the  
244 years 2010-2018 were differing significantly, their annual medians ranging from 6.7 to 7.8  
245 (Figure 3).

246 Another basic parameter is electrical conductivity (EC), which in freshwater is affected  
247 primarily by the geology of the area through which the water flows. The Arctic lake types  
248 range from very dilute waters with EC approaching rainwater, to waters concentrated by  
249 evaporation to beyond the salinity of seawater. Within this range, however, the majority of  
250 lakes are relatively diluted, with  $EC < 300 \mu S cm^{-1}$  (Kling, 2009). The EC values detected in  
251 freshwater samples collected in the summer periods of 2010–2018 differed from each other  
252 significantly in some years (Figure 3), ranging from 7 to  $147 \mu S cm^{-1}$ . This places the studied  
253 lake system within the spectrum of typical Arctic lakes. Between the two lakes which form



254 the studied system, there was a significant increase in EC noted at the transition from upper to  
255 lower (and larger) lake (Wilcoxon signed-rank test  $p < 0.05$ ). The low EC values may be due  
256 to both the predominant substratum geology with siliceous rocks (of limited solubility in  
257 water) and the underlying permafrost, which isolates surface waters and soils from weathering  
258 interactions with deeper mineral soils and rocks. The unfrozen zone beneath the lake may  
259 extend for many meters, but the impact of weathering in this zone on lake chemistry is almost  
260 entirely unknown. Usually, in the rest of the catchment, weathering reactions are confined to  
261 the very shallow unfrozen layers at the surface (Kosek et al., 2019b; Olichwer et al., 2013).

262 **Figure 3.** Range (whiskers) and quartile distribution (Q1 and Q3 frame the box, Q2 = median  
263 shown by the line inside it) of pH and EC [ $\mu\text{S cm}^{-1}$ ] measurements over the 2010, 2012, and  
264 2014-2018 summer seasons. (R) and (L) denote value plotted on right and left axis,  
265 respectively. Kruskal-Wallis ANOVA results are given in the box at the bottom of the graph.  
266 For a colour figure, please refer to the online version of the article

### 267 3.2.2. Trace elements

268 Metals and metalloids were also determined in the samples collected between 2010 and 2018  
269 in the summer periods. Changes in the median concentrations of the elements Al, As, Ba, Be,  
270 Co, Cr, Cu, Ga, Li, Mn, Ni, Rb, Sr, Tl, U, V, and Zn, in the period 2010 – 2018 are shown in  
271 Figure 4, together with their Kruskal-Wallis ANOVA results for inter-annual differences. Of  
272 these elements, the following showed significant differences between the sampling years  
273 ( $p < 0.05$ ): As, Co, Cr, Li, Mn, Ni, Rb, Sr, and Zn (9 out of 17, which can be interpreted that  
274 the trace metal composition of these waters is not stable in time).

275 **Figure 4.** Concentration levels of trace elements in the collected freshwater samples: box  
276 shows the inter-quartile range (50% of all results, 25% on each side of the median, which is  
277 marked as line inside the box); whiskers encompass full range of results. All Y scale units are



278 [ $\mu\text{g L}^{-1}$ ]. (R) next to an element symbol means it is plotted on the right Y axis, (L) – on the  
279 left. Elements are divided into panels due to their varying range of concentration values.  
280 Kruskal-Wallis ANOVA results are printed within each graph. For a colour version of the  
281 figure, please refer to the online version of the article

282 The Revvatnet lake waters contain abundant metals and metalloids due to the presence of ore-  
283 bearing veins and metamorphic rocks in the surrounding area (Smulikowski, 1965;  
284 Wojciechowski, 1964). This geological substratum is more exposed in the upper parts of the  
285 catchment feeding the lake. Furthermore, the spatial variability of the underlying rocks in the  
286 studied catchment allows for the more abundant occurrence of titanium, possibly also barium,  
287 caesium, lithium, rubidium, and zinc in the upper part of the lake waters, of zirconium in the  
288 left tributaries of the middle part, and of chromium and vanadium in both these areas. The  
289 local rocks are relatively abundant in aluminium and manganese throughout the catchment  
290 (Smulikowski, 1965). As for the ore-bearing veins, in the area occur those with chalcopyrite,  
291 cuprite, malachite and azurite, which are copper minerals, as well as smaller concentrations of  
292 sphalerite (with zinc) and galena (with lead). The specific locations of these veins favour the  
293 occurrence of copper near Skoddefjellet mountain and in the left tributaries of the lake, and of  
294 both lead and zinc in the left tributaries of the smallest upper part of the Revvatnet lake  
295 (Wojciechowski, 1964).

296 These elements (except Cs, Pb and Zr, for which we had no or not sufficient data) were tested  
297 with the Wilcoxon signed-rank test between points Rev 1 and Rev 2 (the closest in the two  
298 lakes) and Rev 1 and Rev 5 (top and bottom of the lake system) to find if their concentration  
299 significantly changed between the small lake and the large one. Significant differences (at  $p$   
300  $<0.05$ ) were found between the concentrations of Ba, Cu, Li and Zn in the small lake (Rev 1)  
301 and the output from the large lake (Rev 5); for Al, Ba and Li, the difference was significant  
302 between Rev 1 and Rev 2. Among other elements, with less clear geological divisions in the

303 catchment, statistically significant differences (at  $p>0.05$ ) were found between Rev 1 and Rev  
304 2 for As and Sr, and between top and bottom of the lake system (Rev 1 and Rev 5) for Ga and  
305 Ni. All the noted significant differences were downstream increases, except Zn – it could be  
306 interpreted than only in the case of Zn the local input from ore-bearing veins was more  
307 prominent than the slow dissolution processes of suspended particles which would normally  
308 increase downstream concentrations. This is an opposite pattern to the one noted for lake  
309 bottom sediment upper layer in another flow-through lake system of the temperate zone  
310 (Kuriata-Potasznik et al., 2016), which highlights that different processes govern lake water  
311 and sediment chemistry, therefore direct comparisons may be misleading.

312 Slight changes in the individual concentration levels of metals and metalloids could have been  
313 caused by various intensification of geological processes related to the temperature changes  
314 and to the mineral surface reactions, increased or reduced amount of precipitation in a  
315 selected summer sampling period, complexation, chemical weathering and sorption to solid-  
316 phase soil organic matter (Colombo et al., 2018), as well as due to the variability of  
317 hydrological processes occurring in the research area. Moreover, the elemental concentration  
318 variations may be caused by the occurrence of groundwater associated with the active layer of  
319 permafrost, which gains more importance in the hydrological regime of the Revvatnet lake as  
320 snow patches disappear in the catchment. Apart from the local natural occurrence of metals  
321 and metalloids, they are assumed to be derived to the Arctic mostly from long-range  
322 atmospheric transport (AMAP, 2009), and we consider this issue further in the Discussion  
323 section 4.3.

### 324 3.2.3. Organic compounds as water pollution of the Revvatnet lake

325 Among the chemical characteristics of the Revvatnet lake, the presence of organic compounds  
326 contaminating the study area should also be emphasized. The Arctic is no longer considered a  
327 highly pristine environment, although the air, water, soil and sediment concentrations reveal

328 considerably lower levels of contaminants compared to those found in temperate regions  
329 (Kallenborn et al., 2012). Despite this, some characteristic features of the Arctic, e.g. low  
330 temperatures, precipitation, ice coverage, and extended periods of darkness during winter,  
331 mean that it has the potential to accumulate certain contaminants including persistent organic  
332 pollutants (POPs) in all components of the environment (Hung et al., 2010). There are only a  
333 few local sources of contaminants in the Arctic, such as military installations, industrial  
334 outlets and waste from the old mines, settlements, airports and ships. However, the majority  
335 of Arctic pollution problem has arisen through a combination of long-range transport of  
336 pollutants and the Arctic haze phenomenon, locking the contaminated air in the area for  
337 months (Kallenborn et al., 2012). The cold condensation of POPs enhances the problem of  
338 their deposition in the Arctic (Mackay and Wania, 1995).

339 The Arctic contamination research has mainly focused on POPs (including the PAHs  
340 described in this paper) because they are bioaccumulative, resistant to degradation and toxic.  
341 In addition, other organic chemicals (sum of phenols and formaldehyde) were also determined  
342 in collected freshwater samples, which could testify to their global impact (if they are  
343 confirmed as anthropogenic). Figure 5 shows the average concentration levels of the sum of  
344 phenols, formaldehyde, and total organic carbon (TOC) in the freshwater samples collected  
345 between 2010 and 2018.

346 **Figure 5.** Average concentration levels of the sum of phenols, formaldehyde and total organic  
347 carbon in the collected freshwater samples. For a colour figure, please refer to the online  
348 version of the article

349  $\Sigma$ Phenols and formaldehyde concentrations levels showed no trend over the studied years  
350 (Figure 5), ranging from  $<0.025$  to  $0.261 \text{ mg L}^{-1}$  and from  $<0.005$  to  $0.60 \text{ mg L}^{-1}$ ,  
351 respectively. However, in the case of HCHO, inter-annual variability was significant (as



352 expressed by Kruskal-Wallis ANOVA  $p = 0.00005$ ). For HCHO, out of 55 measurements,  
353 there were two exceptionally high values noted above  $0.30 \text{ mg L}^{-1}$  ( $0.60 \text{ mg L}^{-1}$  at Rev 1 in  
354 July 2012 and  $0.53 \text{ mg L}^{-1}$  at Rev 5 in June 2016), which could impact the interannual  
355 variability picture.  $\Sigma$ Phenols maximum was reached at Rev 2 in September 2015, and  
356 Kruskal-Wallis ANOVA for interannual changes was not significant. The maxima were  
357 formed by rare departures from the much lower concentration levels occurring most  
358 frequently, leading to more uncertainty in estimating annual means than in the case of TOC,  
359 whose concentrations were more uniform, ranging from  $0.45$  to  $1.95 \text{ mg L}^{-1}$ . The interannual  
360 variability of TOC concentrations was, nevertheless, statistically significant (Kruskal-Wallis  
361 ANOVA  $p = 0.0065$ ). None of the three mentioned parameters has differed significantly  
362 between the upper and the lower lake (as measured by the Wilcoxon signed-rank test, run in  
363 two sets of paired samples from: Rev 1 - Rev 2 and Rev 1 - Rev 5).

364 Polycyclic aromatic hydrocarbons are another group of compounds found in the waters of the  
365 Revvatnet lake in the years 2010 – 2018. The distribution of concentrations of the detected  
366 PAHs in the collected lake water samples are shown in Figure 6.

367 **Figure 6.** Concentration levels of determined PAHs in the collected lake water samples: a)  
368 percentage contributions of particular PAHs: b) box and whisker plot, showing median  
369 (middle line), inter-quartile range (box) and full range of results (whiskers). All PAHs  
370 concentrations in  $[\text{ng L}^{-1}]$ . For a colour figure, please refer to the online version of the article

371 Historically, only 16 PAHs have been prioritized as environmentally significant and thus  
372 received the focus of research, however, it is now recognised that aquatic ecosystems may be  
373 exposed to, and potentially affected by hundreds of PAHs and the risks related to that are  
374 poorly understood. From a mixture of 16 PAHs that were analysed, only those that were  
375 detected in the collected samples are shown in Figure 6. Three compounds from the PAH  
376 group occurred in the analysed samples at the highest concentration levels in each year:



377 naphthalene (range of results 69 – 451 ng L<sup>-1</sup>; median 223 ng L<sup>-1</sup>), anthracene (range 22 – 657  
378 ng L<sup>-1</sup>; median 147 ng L<sup>-1</sup>) and fluoranthene (range 13 – 977 ng L<sup>-1</sup>; median 80 ng L<sup>-1</sup>). In the  
379 period 2015 – 2018, these compounds were at similar concentration levels, with no significant  
380 differences in single PAH concentrations between the years (measured with Kruskal-Wallis  
381 ANOVA).

382 The two parts of the studied lake system differed significantly only with respect to some  
383 PAHs, and the changes were not uniform. Wilcoxon signed-rank test yielded significant  
384 results at  $p < 0.05$  for naphthalene and fluorene for the transition from upper to lower lake  
385 (Rev 1 to Rev 2), while acenaphthene and phenanthrene differed significantly in  
386 concentrations at the top and bottom of the lake system (Rev 1 to Rev 5). No other PAHs  
387 showed a consistent change between those points in the sampling scheme.

388 The  $\Sigma$ PAHs concentrations ranged from <MDL to 2151 ng L<sup>-1</sup> in the collected samples, with  
389 annual median concentrations between 303 and 956 ng L<sup>-1</sup> (median concentration in the whole  
390 study period was 565, and average concentration 748 ng L<sup>-1</sup>). A typical distribution of a single  
391 PAH compound concentrations was right-skewed, with the skewness being stronger for PAHs  
392 with a higher number of aromatic rings. With such data distribution, there may be locally or  
393 temporally enhanced pollution effects due to the occurrence of strong maxima in PAHs  
394 concentrations. The pattern and frequency of their occurrence should be subject of a separate  
395 study to establish the environmental risk connected to them.

396 Of interest is the origin of PAHs compounds in the studied remote catchment, and this has  
397 been investigated here using the diagnostic PAHs concentration ratios (Table S2,  
398 Supplementary information). They show a uniformly pyrogenic origin of the detected PAHs.  
399 In no sample, the petrogenic ratios of ANT/(ANT+PHE) < 0.1 or FLA/(FLA+PYR) < 0.4  
400 have been found (De La Torre-Roche et al., 2009; Pies et al., 2008). Based on the  
401 FLA/(FLA+PYR) and FL/(FL+PYR) ratios, further distinctions can be made. The majority of



402 the collected samples exhibited both ratios exceeding 0.5, which for the latter is interpreted as  
403 indicating petrol combustion origin (Ravindra et al., 2006), while the former suggests an  
404 origin from grass, wood, and coal combustion (De La Torre-Roche et al., 2009) or diesel  
405 (Ravindra et al., 2008). Thus, a combination of various combustion processes is likely to have  
406 produced the local PAHs composition, especially as there are stable and low local sources of  
407 diesel combustion to fuel the station (there is also a waste incinerator and multiple vehicles  
408 necessary for the operation of the Polish Polar Station), less stable regional sources, e.g. ships  
409 en route near Svalbard, and the long-range transport of both fossil-fuel- and biomass-burning-  
410 related PAHs (Granberg et al., 2017; Winther et al., 2014; Zielinski et al., 2020). The lower  
411 ratios of FLA/(FLA+PYR) can be interpreted as fossil fuel combustion (De La Torre-Roche  
412 et al., 2009), and FL/(FL+PYR) < 0.5 may come from diesel use (Ravindra et al., 2006).  
413 These have occurred first in 2010, then in September 2015 and 2016 only in the smaller upper  
414 lake (Rev 1 location), and only in 2018 they occurred in more samples again. In the context of  
415 the location of the small lake (more distant from the local human activity in the polar station)  
416 and these ratios occurring more often in September, they can be treated as the background  
417 pollution from long-range transport and remobilized from the melting permafrost. On the  
418 other hand, the higher ratios may show a combination of more frequent biomass burning  
419 influence and the local fuel exhaust being carried in certain meteorological conditions into the  
420 Revelva valley. The strongest conclusion from the calculation of these indicator ratios is the  
421 lack of petrogenic source influence in the area, which is consistent with its geological  
422 substratum lacking bituminous material.

423

#### 424 **4. Discussion**

##### 425 4.1. Interactions between various chemical factors – principal component analysis



426 The overview of the chemical parameters in the sampling points of the Revvatnet (Figure 7a)  
427 highlights the main chemical division between parts of the summer season when samples have  
428 been collected. The particular sampling locations did not show any striking differences within  
429 the collected dataset – it should be especially noted that the overall composition of the upper,  
430 smaller lake at Rev 1 is similar to the chemical parameters of the large Revvatnet proper. The  
431 year 2015 differed from the later period, yet this may represent also the different month of  
432 sample collection within that year (July instead of June representing the early summer). The  
433 highest contribution to overall variability in the dataset was contributed by several metals and  
434 metalloids: Al, B, Ba, Ga, Li, Mn, Ni and Sr (Figure 7b), as well as TOC.

435 **Figure 7.** Principal component analysis, exploring the variability in the entire collected  
436 chemical dataset in the space defined by two most prominent components (representing >37%  
437 of total variability in the dataset): a.) & b.) graph for individual datapoints, with the month  
438 and sampling site highlighted by colour in a.) and b.), respectively; c.) graph for variables; For  
439 a colour figure, please refer to the online version of the article

440 PCAs were also conducted for subsets of variables of similar character, to explore the possible  
441 reduction of the dataset (or to exhibit the variables influencing the differences in the dataset  
442 the most). For metals and metalloids, if treated separately, the main variability drivers were:  
443 Al, B, Ba, Li, Sr, but also V and Cr. They effectively separated the data points by both  
444 sampling month and year (except the 2016-2018 period, which remained relatively similar)  
445 (Figure S2, Supplementary information). The variability was spread rather evenly in the  
446 PAHs domain, and those could be divided into two closely correlated groups: 1) phenanthrene  
447 and pyrene, which shared some variability in concentrations with TOC, 2) the other six  
448 frequently detected compounds, changing independently from TOC concentrations. A  
449 significant proportion of the variability in PAHs concentrations (approximated by PC2 in  
450 Figure S3b&c) appears to have resulted from an intra-seasonal difference in hydrochemical

451 regime (e.g. snowmelt as opposed to permafrost thaw as a source of PAHs in the collected  
452 samples). PC1 in Figure S3c, representing 49% of the variability, has been compromised by a  
453 clear outlier collected in 2010, which makes it more difficult to interpret. A renewed analysis  
454 without the outlier yielded a clearer seasonal division (Figure S3a&b), where PHE and PYR  
455 high concentrations aligned with late summer sampling time, while NAP and ACE did so in  
456 the early summer samples. The site Rev 5 was also more clearly distinguished by its higher  
457 contribution to total variability of the concentrations of PAHs (Figure S3b). Notably, in point  
458 Rev 1 occurred the maxima in ANT concentrations, while in Rev 5 the absolute maxima for  
459 any single PAH concentration were found (among all sampling points), and these extreme  
460 values concerned FLA. Those resulted also in the highest  $\Sigma$ PAHs found at that location. Thus,  
461 it appears that the flow of water through the lake does not necessarily facilitate cleansing it of  
462 PAHs pollution (Hamilton et al., 2001).

#### 463 4.2. A concise overview of other Arctic lake systems research

464 In the Arctic, the problem of the appearance of pollutants in various elements of the  
465 environment is widespread, and therefore it is widely studied by scientists from around the  
466 world. Researching lakes and river catchments in terms of their chemical composition, as well  
467 as hydrochemical parameters of the catchments, is increasingly the subject of interest of  
468 researchers throughout Spitsbergen and other parts of the Arctic (e.g., Franczak et al., 2016;  
469 Lehmann-Konera et al., 2019; Mazurek et al., 2012; Szumińska et al., 2018; Wawrzyniak et  
470 al., 2020). Lakes in the Arctic are typically ultra-oligotrophic and fed by allochthonous  
471 nutrients (Hamilton et al., 2001; Lim and Douglas, 2003).

472 The most frequently studied areas in Spitsbergen in terms of the hydrochemistry of lakes are  
473 the areas adjacent to the fjord Hornsund, Bellsund, Isjord and Kongsfjord, which are strongly  
474 influenced by marine aerosols, which may introduce several metals and metalloids into those  
475 catchments (e.g., Nawrot et al., 2016). The content of the metals such as Sr or Ba was at a

476 similar level in Revvatnet as in the other areas of Svalbard (Szumińska et al., 2018), and Sr  
477 was mentioned there to come from geogenic sources in the lakes located on a raised marine  
478 terrace, which is a similar feature to the environment surrounding the Revvatnet. The general  
479 levels of metal and metalloid concentrations in Revvatner were also comparable to the state of  
480 Lake Imandra (in the Russian Arctic) prior to the intensive human influence in the  
481 Monchegorsk area (in the 1930s; Moiseenko et al., 2009), highlighting the representativeness  
482 of our study site in terms of general hydrochemical features of Arctic lakes. Similarly to the  
483 study results from Canadian Arctic (Lim and Douglas, 2003), in Revvatnet, several metals  
484 and metalloids concentrations were also influenced by the local geology. From early to late  
485 summer, most metal concentrations in Revvatnet increased or remained similar, consistently  
486 with a study on seasonal changes in thermokarst lakes by Manasypov et al. (2015).

487 The organic compounds content, including pollutants, has also been tested in many  
488 catchments in Spitsbergen and elsewhere in the Arctic. Smaller lakes tend to have higher TOC  
489 concentrations (Lim and Douglas, 2003). Similar levels of PAHs concentrations were  
490 determined, for example, in the nearby Fuglebekken catchment (average sum of determined  
491 PAHs ranged from 4-600 ng L<sup>-1</sup> (Kozak et al., 2017). The content of PAHs as well as other  
492 pollutants, e.g. phenols and aldehydes, as well as microplastic, was determined in lakes in  
493 Bellsund area (Lehmann-Konera et al., 2019, 2018) and those located in Ny-Ålesund area  
494 (González-Pleiter et al., 2020; Jiao et al., 2009). Persistent organic pollutants have also been  
495 detected extensively in two high Arctic lakes in Bjørnøya (Bear Island) (Evenset et al., 2004) .  
496 As the lakes on Svalbard are remote, more than hundreds km from any known point source,  
497 the presence of POPs suggests that they have been transported to the area mainly by long-  
498 range transport (Evenset et al., 2004), including through the pathway of seabird guano  
499 (Evenset et al., 2007). As POPs pose a serious threat to the entire polar ecosystem, their  
500 redistribution in the dynamic environment of the Arctic and the combined effects of pollution

501 and climate change warrant further investigation in the times of increased strain on the Arctic  
502 environment (AMAP, 2011).

503 PAHs are typically formed during the incomplete combustion of fossil fuels, biomass, and  
504 through other industrial activities; they also occur naturally in bituminous rocks. They have  
505 been found in the Arctic environment, originating both from the long-range atmospheric  
506 transport (Wang et al., 2013) and the local sources. PAHs have been found widely in polar  
507 environmental media: the atmosphere, water, sediments and biota (Kosek et al., 2019a, 2019b,  
508 2018, 2017, 2016; Kozak et al., 2017; Koziol et al., 2020; Polkowska et al., 2011; Potapowicz  
509 et al., 2019).

510 PAHs are generally hydrophobic and many interact strongly with sedimentary organic carbon  
511 (Burgess et al., 2003) and as such, PAHs are commonly associated with sediments and  
512 particulate matter, which plays a role in their removal from lake water. They also  
513 bioaccumulate in aquatic biota, particularly with the lower trophic levels (Besten et al., 2003;  
514 Kosek et al., 2019b; Neff et al., 2005), and the ecotoxicological concerns have focused on  
515 those. The toxicology of PAHs in the aquatic environments has been well documented and  
516 numerous articles/books are available in relation to bioavailability (Besten et al., 2003;  
517 Burgess et al., 2003), bioaccumulation (Besten et al., 2003; Meador, 2003) and toxicity  
518 (Albers, 2003; Logan, 2007; Malcolm and Shore, 2003).

519

520 4.3. Revvatnet lake system in comparison to the environmental concentrations of pollutants in  
521 the Hornsund area

522 As the described flow-through lake system is part of larger landscape, where pollution has  
523 been studied by other researchers, we discuss its contamination level also in the context of  
524 linkages to other environmental media in the surroundings. While it is impossible to compare  
525 directly the metal concentrations in lake water to the concentrations in sea sediment, it is

526 nevertheless valuable information to see whether the metals with elevated concentration in the  
527 catchment of the Revvatnet lake are connected to elevated concentrations of the same metals  
528 in sediments located downstream from such a catchment. In Zaborska et al. (2017) the Pb  
529 concentration levels found in the outside part of the Hornsund fjord (near Revelva catchment)  
530 were elevated against the levels further inside the fjord (unlike the levels of Cu, Cd or Zn,  
531 which were in a similar range for those sites). Of these four metals, we have complete data  
532 only in 2010, later on only Zn and Cu concentrations have been measured. However, the  
533 measured concentration at a ppb level or lower in lake water is not likely to influence the  
534 variability in marine sediment concentrations, which range from 0.01 to >100 ppm. This is  
535 consistent with the conclusion by Zaborska et al. (2017), that large-scale atmospheric and  
536 oceanic processes influence the concentrations in ocean sediment, and not the local runoff. A  
537 similar comparative material, but for PAHs, can be found in Pouch et al. (2017) article on  
538 fjord sediments from Svalbard. The concentrations measured by them ranged 37.3 to 1973  
539 ppb for  $\Sigma 12$  PAHs. Such levels are more likely to be influenced by the local runoff, however  
540 such an influence is far from certain (the concentrations in lake water oscillated around 1 ppb  
541 for  $\Sigma 8$  PAHs). This is consistent with the interpretation of spatial variability in POPs  
542 concentrations in the fjord sediments may be influenced by local secondary sources of  
543 contamination, such as melting glaciers, especially as sediment is a preferential deposition  
544 media for POPs (in comparison to water) due to their hydrophobicity. The sources of PAHs in  
545 both lake water and sea sediment, according to PAHs concentration indicator ratios, were  
546 consistently pyrolytic (Table S2 and Pouch et al., 2017). We conclude that the catchments,  
547 which are in majority not glaciated, like that of the Revvatnet lake, also contain non-  
548 negligible POPs contamination and should be monitored for their concentration levels in the  
549 future.



550 A high concentration of the  $^{90}\text{Sr}$  radionuclide, which is of anthropogenic descent, was found  
551 on the nearby Hans glacier (Łokas et al., 2016), indicating that perhaps the high  
552 concentrations of Sr in the waters of Revvatnet (up to 80 ppb) were also influenced by human  
553 activity (possibly via long-range transport of contamination). Furthermore, the cryoconite  
554 upon Hans glacier was found to have heavy metal concentrations beyond those noted in local  
555 rocks, especially for Pb, Cd, Cu, Zn, which testifies to their enhanced deposition from long-  
556 range transport or more efficient accumulation in cryoconite holes than elsewhere. While the  
557 comparison between water and sediment concentrations is not directly possible, we observe  
558 that Zn and Cu were important components of the metal impurity concentrations present in  
559 the Revvatnet waters (while Pb and Cd were only measured in 2010, and therefore they  
560 cannot be included in this comparison). This observation cautions that at least part of the  
561 metal load found in the Revvatnet may be anthropogenic, including long-range transported  
562 contamination. However, the concentrations of these metals did not correlate with the  
563 anthropogenic radionuclide concentrations in Łokas et al. (2016) study.

564 Since the origin of metals in cryoconite is most likely from atmospheric deposition, this  
565 finding is consistent with Kozak et al. (2015), who found Zn and Cu to be provided in excess  
566 by atmospheric precipitation, and likely from anthropogenic contamination. However, river  
567 and lake waters experience longer contact with the geological substratum, and normally they  
568 note higher concentration of soluble components present in parent rock of the area. With  
569 respect to precipitation, it is also helpful to explore whether precipitation events bring a  
570 significant load of certain metals, which has been shown to be possible especially for Mn, Sr  
571 and Zn in 2010 (Kozioł et al., 2020). Furthermore, Sr was found to show a higher  
572 concentration in smaller lakes of the area than in Revvatnet (in 2010; Kozioł et al., 2020),  
573 which supports it could have originated from precipitation. In Kozak et al. (2015) study on  
574 precipitation waters in the area, Mn and Sr were notably changing in concentration, depending



575 on the local wind direction during the rainfall event, which could mean that despite their  
576 transfer through rain, these metals have local rather than distant sources in the Hornsund  
577 region. Furthermore, in the snow cover of Hans glacier (Koziol et al., 2021; *in press*), both  
578 Mn and Sr were found to cluster (by concentration variability) with natural sources in rock  
579 and seawater, respectively, and Sr was shown to originate from seawater in 39% (median  
580 seawater fraction of Sr in the snow samples collected). On the other hand, Szumińska et al.,  
581 (2018) claim that Sr in lakes in Bellsund area is geogenic, while Mn, Co, Ni, Cu, Ga, Ba and  
582 Cd in that site are interpreted to be transported there airborne. In the Koziol et al. (2021; *in*  
583 *press*) study, Cu also clustered with the natural geogenic elements, as did Mn. In the same  
584 study, Zn clustered with the likely antropogenic elements from long-range transport.  
585 However, due to its presence in ore-bearing veins in the Revvatnet surroundings, the  
586 composition of surface waters may still be impacted importantly by substratum, and not only  
587 precipitation, and this is supported by the spatial distribution of Zn concentrations in the lakes  
588 of the area (Koziol et al., 2020).

589

## 590 **5. Summary and conclusions**

591 The particular chemical compositions of Arctic lakes are dependent on several factors,  
592 including proximity to the ocean and geology for inorganic materials, and for organic  
593 compounds compositions depend on the extent and type of terrestrial vegetation in the  
594 surrounding catchment, as well as anthropogenic pollution (ACIA, 2005; AMAP, 2016;  
595 Kling, 2009). In this article, we presented the characteristics of one such a lake system, the  
596 flow-through Revvatnet in southern Svalbard, composed of a smaller upper lake and a large  
597 lower lake, cone-shaped, reaching -29.1 m depth. The system has typical morphometric  
598 characteristics of a lake formed by glacial processes. The pH of its waters was near-neutral, its  
599 annual medians ranging 6.7 to 7.8, while the EC was rather low (which is typical for many

600 Arctic lakes), ranging 7 to 147  $\mu\text{S cm}^{-1}$ . The significant downstream changes in metal and  
601 metalloid concentrations consisted of increases downstream (with the exception of Zn), which  
602 is likely a result of the slow dissolution of suspended particles in the lake system. However,  
603 such changes for PAHs were not consistent in direction and relatively few. The  $\Sigma\text{PAHs}$   
604 concentrations ranged from <LOD to 2151  $\text{ng L}^{-1}$ , and they were not petrogenic according to  
605 indicator PAHs concentration ratios – they appear to come from a mixture of combustion  
606 processes, near and far, from fossil fuel and biomass. The chemical composition of these  
607 waters was characterised by a strong seasonal variability, and in terms of metals and  
608 metalloids especially – also significant inter-annual variability. Of particular concern in terms  
609 of pollution in these waters are the high maxima in organic compound and metal or metalloid  
610 concentrations, and the patterns of their occurrence requires further study to determine the  
611 environmental risk related to them.

612

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622

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626

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