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

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

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

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

1. Introduction

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

High-latitude lakes are likely to show ongoing responses to present climate instability that leads to deglaciation, variations in chemical composition of freshwater, and, for lakes located close to coastal waters, changes in their linkages with the sea. All of these processes are major controls on the structure and functioning of aquatic ecosystems (Van Hove et al., 2006; Vincent and Laybourn-Parry, 2008). Geological variety of the polar regions, including the differences in substrate lithology, may also affect the extent of rock weathering and the chemical composition of soil water that finally discharges into polar lakes (Hamilton et al., 2001; Szumińska et al., 2018; Vincent and Laybourn-Parry, 2008). In brief, Arctic lake ecosystems are strongly coupled to the features of their surrounding landscapes, such as hydrological characteristics, geomorphology, lithology and vegetation, and due to this, there are several significant differences in lake characteristics in various parts of the Arctic (Wetzel, 2001).

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

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

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

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

2. Materials and Methods

2.1. Study area

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

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

2.2. Bathymetric measurements

Bathymetric measurements were collected in August 2010 using a Lowrance LCX-17MT echo sounder system with a teleCover-Lok telescopic handle mounted on the transom of the rubber boat, recording the path of the boat movement with an integrated GPS receiver (LGC-2000). Bathymetric measurements were made according to the methodology proposed by Lange (1993) and presented in the guide for hydrographic fieldwork (Gutry-Korycka and Werner-Więckowska, 1996). The development of the bathymetric plan of the lake required a series of cross-sections and longitudinal sections, and then the transfer of information about the distribution of depth in these profiles to the digital map of the lake. The total length of all bathymetric profiles was over 20 km (2700 measuring points), which allowed to draw up a bathymetric plan as well as a three-dimensional model of the lake basin. ArcGIS 10.6.1 (ESRI) and Surfer 18 softwares (Golden Software) were used to make the bathymetric plan and the lake model.

2.3. Freshwater sampling

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

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

Samples were collected manually from the Revvatnet lake at a distance of 1.5 m from the shore with no headspace into air-tight, chemically clean bottles (the purity of the procedure has been verified by daily blank sample collection). Pre-cleaning procedure for the bottles included week-long soaking with Milli-Q deionised water and removing the water from the sampling containers several times. The water was sampled from a depth between 20 and 50 cm below water level.

2.4. Chemical analyses

Milli-Q deionised water (18 MΩ) was used during the determination of the various target analyte groups to determine instrumental background (by inserting it in the analysis queue once every six samples) and to prepare sample container blanks. The concentrations of chemical compounds in the collected samples were determined by: 1) for metals and metalloids: Inductively Coupled Plasma – Mass Spectrometry (the element concentration CVs of the obtained triplicate results ranged from 0.5 to 1.5%); 2) for total organic carbon (TOC): TOC-VCSH/CSN Analyser (Shimadzu, Japan) with NDIR detector; 3) for formaldehyde and the sum of phenols: a spectrophotometer, and 4) for PAHs: a gas chromatograph coupled toa single quadrupole mass spectrometer. All the applied analytical procedures have been validated against certified reference materials (CRMs) concordant with ISO Guide 34:2009 and ISO/IEC 17025:2005, and the data obtained here were subject to strict QC procedures. Prior to pH measurements, a three-point electrode calibration was performed with temperature compensation, using MERCK Millipore Certipur® buffer solutions of pH 4.00, 7.00 and 9.00 (at 25°C). In the analysis of metal and metalloid concentrations, we applied the Standard Reference Material (RM) NIST 1643e Trace Elements in Water, and RM Enviro MAT ES-L-2CRM, ES-H-2 CRM SCP SCIENCE. The calibration of the apparatus was based on RMs by Inorganic ventures ANALITYK: CCS-4, CCS-6, CCS-1, IV-ICPMS-71A. Potassium hydrogen phthalate by NacalaiTesque (Japan) was used for the calibration of the TOC Analyser. Each sample was analysed in triplicate. Sensitivity tests were performed by injecting standard analyte mixtures within the measured concentration range, and linear calibration curves of the peak area against concentration showed correlation coefficients (R^2) in the range of 0.898–0.999 for all standards. Detailed technical specifications of the applied determination techniques, as well as the validation parameters of the analytical procedures, are summarised in Table S1 (Supplementary information).

2.5. Statistical analyses

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

Principal Component Analysis (PCA) is a multivariate statistical analysis that allows revealing internal relations in the data set. PCA finds linear combinations of the original variables, referred to as principal components, which provide better descriptors of the data pattern than the original measurements and account for most of the dataset variation. Principal component analysis (PCA) has been conducted in R software (version 3.6.2.), using the *FactoMineR* package. Missing data were ommitted by case. Prior to analysis, values <MDL have been replaced with ½ of the MDL value, subsequently all variables except pH (which already is a logarithm) were log-transformed to bring their distribution closer to normal. Departures from normality were the most pronounced where there were several <MDL values. Due to the exploratory character of the performed PCA, we accepted minor departures from the normal distribution without further data transformation. All variables were scaled (i.e. z-transformed) as part of the PCA calculation procedure (by using the parameter scale.unit=TRUE). R was also used to calculate the Wilcoxon signed-rank test results (with the *wilcox.test* function) for the matched samples (i.e. from two sites in the same lake system, which are connected by water flow).

3. Results

3.1. Hydrological characteristics of the lake (morphometry)

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

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

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

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

The chemistry of polar freshwater reservoirs is shaped by the geological substratum, as well as by atmospheric deposition, marine aerosols, chemical weathering, biological processes, glacial and periglacial activity. Depending on the geological structures and soil cover on the raised marine terraces and in the surrounding areas, the waters that reach tundra lakes transport varying proportions of specific chemicals (Mazurek et al., 2012). The results obtained for the Revvatnet samples from the summer periods of 2010 to 2018 are shown in Table S3 (Supplementary information).

3.2.1. pH and electrical conductivity (EC) measurements

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

Another basic parameter is electrical conductivity (EC), which in freshwater is affected primarily by the geology of the area through which the water flows. The Arctic lake types range from very dilute waters with EC approaching rainwater, to waters concentrated by evaporation to beyond the salinity of seawater. Within this range, however, the majority of lakes are relatively diluted, with EC < 300 $\mu S\ cm^{-1}$ (Kling, 2009). The EC values detected in freshwater samples collected in the summer periods of 2010–2018 differed from each other significantly in some years (Figure 3), ranging from 7 to 147 $\mu S\ cm^{-1}$. This places the studied lake system within the spectrum of typical Arctic lakes. Between the two lakes which form the studied system, there was a significant increase in EC noted at the transition from upper to lower (and larger) lake (Wilcoxon signed-rank test $p < 0.05$). The low EC values may be due to both the predominant substratum geology with siliceous rocks (of limited solubility in water) and the underlying permafrost, which isolates surface waters and soils from weathering interactions with deeper mineral soils and rocks. The unfrozen zone beneath the lake may extend for many meters, but the impact of weathering in this zone on lake chemistry is almost entirely unknown. Usually, in the rest of the catchment, weathering reactions are confined to the very shallow unfrozen layers at the surface (Kosek et al., 2019b; Olichwer et al., 2013).

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

3.2.2. Trace elements

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

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

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

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

These elements (except Cs, Pb and Zr, for which we had no or not sufficient data) were tested with the Wilcoxon signed-rank test between points Rev 1 and Rev 2 (the closest in the two lakes) and Rev 1 and Rev 5 (top and bottom of the lake system) to find if their concentration significantly changed between the small lake and the large one. Significant differences (at $p < 0.05$) were found between the concentrations of Ba, Cu, Li and Zn in the small lake (Rev 1) and the output from the large lake (Rev 5); for Al, Ba and Li, the difference was significant between Rev 1 and Rev 2. Among other elements, with less clear geological divisions in the catchment, statistically significant differences (at $p>0.05$) were found between Rev 1 and Rev 2 for As and Sr, and between top and bottom of the lake system (Rev 1 and Rev 5) for Ga and Ni. All the noted significant differences were downstream increases, except Zn – it could be interpreted than only in the case of Zn the local input from ore-bearing veins was more prominent than the slow dissolution processes of suspended particles which would normally increase downstream concentrations. This is an opposite pattern to the one noted for lake bottom sediment upper layer in another flow-through lake system of the temperate zone (Kuriata-Potasznik et al., 2016), which highlights that different processes govern lake water and sediment chemistry, therefore direct comparisons may be misleading.

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

3.2.3. Organic compounds as water pollution of the Revvatnet lake

Among the chemical characteristics of the Revvatnet lake, the presence of organic compounds contaminating the study area should also be emphasized. The Arctic is no longer considered a highly pristine environment, although the air, water, soil and sediment concentrations reveal considerably lower levels of contaminants compared to those found in temperate regions (Kallenborn et al., 2012). Despite this, some characteristic features of the Arctic, e.g. low temperatures, precipitation, ice coverage, and extended periods of darkness during winter, mean that is has the potential to accumulate certain contaminants including persistent organic pollutants (POPs) in all components of the environment (Hung et al., 2010). There are only a few local sources of contaminants in the Arctic, such as military installations, industrial outlets and waste from the old mines, settlements, airports and ships. However, the majority of Arctic pollution problem has arisen through a combination of long-range transport of pollutants and the Arctic haze phenomenon, locking the contaminated air in the area for months (Kallenborn et al., 2012). The cold condensation of POPs enhances the problem of their deposition in the Arctic (Mackay and Wania, 1995).

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

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

∑Phenols and formaldehyde concentrations levels showed no trend over the studied years (Figure 5), ranging from <0.025 to 0.261 mg L^{-1} and from <0.005 to 0.60 mg L^{-1}, respectively. However, in the case of HCHO, inter-annual variability was significant (as expressed by Kruskal-Wallis ANOVA p = 0.00005). For HCHO, out of 55 measurements, there were two exceptionally high values noted above 0.30 mg L^{-1} (0.60 mg L^{-1} at Rev 1 in July 2012 and 0.53 mg L^{-1} at Rev 5 in June 2016), which could impact the interannual variability picture. ∑Phenols maximum was reached at Rev 2 in September 2015, and Kruskal-Wallis ANOVA for interannual changes was not significant. The maxima were formed by rare departures from the much lower concentration levels occurring most frequently, leading to more uncertainty in estimating annual means than in the case of TOC, whose concentrations were more uniform, ranging from 0.45 to 1.95 mg L^{-1}. The interannual variability of TOC concentrations was, nevertheless, statistically significant (Kruskal-Wallis ANOVA p = 0.0065). None of the three mentioned parameters has differed significantly between the upper and the lower lake (as measured by the Wilcoxon signed-rank test, run in two sets of paired samples from: Rev 1 - Rev 2 and Rev 1 - Rev 5).

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

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

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

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

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

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

Of interest is the origin of PAHs compounds in the studied remote catchment, and this has been investigated here using the diagnostic PAHs concentration ratios (Table S2, Supplementary information). They show a uniformly pyrogenic origin of the detected PAHs. In no sample, the petrogenic ratios of ANT/(ANT+PHE) < 0.1 or FLA/(FLA+PYR) < 0.4 have been found (De La Torre-Roche et al., 2009; Pies et al., 2008). Based on the FLA/(FLA+PYR) and FL/(FL+PYR) ratios, further distinctions can be made. The majority of the collected samples exhibited both ratios exceeding 0.5, which for the latter is interpreted as indicating petrol combustion origin (Ravindra et al., 2006), while the former suggests an origin from grass, wood, and coal combustion (De La Torre-Roche et al., 2009) or diesel (Ravindra et al., 2008). Thus, a combination of various combustion processes is likely to have produced the local PAHs composition, especially as there are stable and low local sources of diesel combustion to fuel the station (there is also a waste incinerator and multiple vehicles necessary for the operation of the Polish Polar Station), less stable regional sources, e.g. ships en route near Svalbard, and the long-range transport of both fossil-fuel- and biomass-burning-related PAHs (Granberg et al., 2017; Winther et al., 2014; Zielinski et al., 2020). The lower ratios of FLA/(FLA+PYR) can be interpreted as fossil fuel combustion (De La Torre-Roche et al., 2009), and FL/(FL+PYR) < 0.5 may come from diesel use (Ravindra et al., 2006). These have occurred first in 2010, then in September 2015 and 2016 only in the smaller upper lake (Rev 1 location), and only in 2018 they occurred in more samples again. In the context of the location of the small lake (more distant from the local human activity in the polar station) and these ratios occurring more often in September, they can be treated as the background pollution from long-range transport and remobilized from the melting permafrost. On the other hand, the higher ratios may show a combination of more frequent biomass burning influence and the local fuel exhaust being carried in certain meteorological conditions into the Revelva valley. The strongest conclusion from the calculation of these indicator ratios is the lack of petrogenic source influence in the area, which is consistent with its geological substratum lacking bituminous material.

4. Discussion

4.1. Interactions between various chemical factors – principal component analysis

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

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

PCAs were also conducted for subsets of variables of similar character, to explore the possible reduction of the dataset (or to exhibit the variables influencing the differences in the dataset the most). For metals and metalloids, if treated separately, the main variability drivers were: Al, B, Ba, Li, Sr, but also V and Cr. They effectively separated the data points by both sampling month and year (except the 2016-2018 period, which remained relatively similar) (Figure S2, Supplementary information). The variability was spread rather evenly in the PAHs domain, and those could be divided into two closely correlated groups: 1) phenanthrene and pyrene, which shared some variability in concentrations with TOC, 2) the other six frequently detected compounds, changing independently from TOC concentrations. A significant proportion of the variability in PAHs concentrations (approximated by PC2 in Figure S3b&c) appears to have resulted from an intra-seasonal difference in hydrochemical regime (e.g. snowmelt as opposed to permafrost thaw as a source of PAHs in the collected samples). PC1 in Figure S3c, representing 49% of the variability, has been compromised by a clear outlier collected in 2010, which makes it more difficult to interpret. A renewed analysis without the outlier yielded a clearer seasonal division (Figure S3a&b), where PHE and PYR high concentrations aligned with late summer sampling time, while NAP and ACE did so in the early summer samples. The site Rev 5 was also more clearly distinguished by its higher contribution to total variability of the concentrations of PAHs (Figure S3b). Notably, in point Rev 1 occurred the maxima in ANT concentrations, while in Rev 5 the absolute maxima for any single PAH concentration were found (among all sampling points), and these extreme values concerned FLA. Those resulted also in the highest ∑PAHs found at that location. Thus, it appears that the flow of water through the lake does not necessarily facilitate cleansing it of PAHs pollution (Hamilton et al., 2001).

4.2. A concise overview of other Arctic lake systems research

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

The most frequently studied areas in Spitsbergen in terms of the hydrochemistry of lakes are the areas adjacent to the fjord Hornsund, Bellsund, Isjord and Kongsfjord, which are strongly influenced by marine aerosols, which may introduce several metals and metalloids into those catchments (e.g., Nawrot et al., 2016). The content of the metals such as Sr or Ba was at a similar level in Revvatnet as in the other areas of Svalbard (Szumińska et al., 2018), and Sr was mentioned there to come from geogenic sources in the lakes located on a raised marine terrace, which is a similar feature to the environment surrounding the Revvatnet. The general levels of metal and metalloid concentrations in Revvatner were also comparable to the state of Lake Imandra (in the Russian Arctic) prior to the intensive human influence in the Monchegorsk area (in the 1930s; Moiseenko et al., 2009), highlighting the representativeness of our study site in terms of general hydrochemical features of Arctic lakes. Similarly to the study results from Canadian Arctic (Lim and Douglas, 2003), in Revvatnet, several metals and metalloids concentrations were also influenced by the local geology. From early to late summer, most metal concentrations in Revvatnet increased or remained similar, consistently with a study on seasonal changes in thermokarst lakes by Manasypov et al. (2015).

The organic compounds content, including pollutants, has also been tested in many catchments in Spitsbergen and elsewhere in the Arctic. Smaller lakes tend to have higher TOC concentrations (Lim and Douglas, 2003). Similar levels of PAHs concentrations were determined, for example, in the nearby Fuglebekken catchment (average sum of determined PAHs ranged from 4-600 ng L^{-1} (Kozak et al., 2017). The content of PAHs as well as other pollutants, e.g. phenols and aldehydes, as well as microplastic, was determined in lakes in Bellsund area (Lehmann-Konera et al., 2019, 2018) and those located in Ny-Ålesund area (González-Pleiter et al., 2020; Jiao et al., 2009). Persistent organic pollutants have also been detected extensively in two high Arctic lakes in Bjørnøya (Bear Island) (Evenset et al., 2004) . As the lakes on Svalbard are remote, more than hundreds km from any known point source, the presence of POPs suggests that they have been transported to the area mainly by long-range transport (Evenset et al., 2004), including through the pathway of seabird guano (Evenset et al., 2007). As POPs pose a serious threat to the entire polar ecosystem, their redistribution in the dynamic environment of the Arctic and the combined effects of pollution and climate change warrant further investigation in the times of increased strain on the Arctic environment (AMAP, 2011).

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

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

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

As the described flow-through lake system is part of larger landscape, where pollution has been studied by other researchers, we discuss its contamination level also in the context of linkages to other environmental media in the surroundings. While it is impossible to compare directly the metal concentrations in lake water to the concentrations in sea sediment, it is nevertheless valuable information to see whether the metals with elevated concentration in the catchment of the Revvatnet lake are connected to elevated concentrations of the same metals in sediments located downstream from such a catchment. In Zaborska et al. (2017) the Pb concentration levels found in the outside part of the Hornsund fjord (near Revelva catchment) were elevated against the levels further inside the fjord (unlike the levels of Cu, Cd or Zn, which were in a similar range for those sites). Of these four metals, we have complete data only in 2010, later on only Zn and Cu concentrations have been measured. However, the measured concentration at a ppb level or lower in lake water is not likely to influence the variability in marine sediment concentrations, which range from 0.01 to >100 ppm. This is consistent with the conclusion by Zaborska et al. (2017), that large-scale atmospheric and oceanic processes influence the concentrations in ocean sediment, and not the local runoff. A similar comparative material, but for PAHs, can be found in Pouch et al. (2017) article on fjord sediments from Svalbard. The concentrations measured by them ranged 37.3 to 1973 ppb for Σ12 PAHs. Such levels are more likely to be influenced by the local runoff, however such an influence is far from certain (the concentrations in lake water oscillated around 1 ppb for Σ8 PAHs). This is consistent with the interpretation of spatial variability in POPs concentrations in the fjord sediments may be influenced by local secondary sources of contamination, such as melting glaciers, especially as sediment is a preferential deposition media for POPs (in comparison to water) due to their hydrophobicity. The sources of PAHs in both lake water and sea sediment, according to PAHs concentration indicator ratios, were consistently pyrolytic (Table S2 and Pouch et al., 2017). We conclude that the catchments, which are in majority not glaciated, like that of the Revvatnet lake, also contain non-negligible POPs contamination and should be monitored for their concentration levels in the future.

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

Since the origin of metals in cryoconite is most likely from atmospheric deposition, this finding is consistent with Kozak et al. (2015), who found Zn and Cu to be provided in excess by atmospheric precipitation, and likely from anthropogenic contamination. However, river and lake waters experience longer contact with the geological substratum, and normally they note higher concentration of soluble components present in parent rock of the area. With respect to precipitation, it is also helpful to explore whether precipitation events bring a significant load of certain metals, which has been shown to be possible especially for Mn, Sr and Zn in 2010 (Koziol et al., 2020). Furthermore, Sr was found to show a higher concentration in smaller lakes of the area than in Revvatnet (in 2010; Koziol et al., 2020), which supports it could have orginated from precipitation. In Kozak et al. (2015) study on precipitation waters in the area, Mn and Sr were notably changing in concentration, depending on the local wind direction during the rainfall event, which could mean that despite their transfer through rain, these metals have local rather than distant sources in the Hornsund region. Furthermore, in the snow cover of Hans glacier (Koziol et al., 2021; *in press*), both Mn and Sr were found to cluster (by concentration variability) with natural sources in rock and seawater, respectively, and Sr was shown to originate from seawater in 39% (median seawater fraction of Sr in the snow samples collected). On the other hand, Szumińska et al., (2018) claim that Sr in lakes in Bellsund area is geogenic, while Mn, Co, Ni, Cu, Ga, Ba and Cd in that site are interpreted to be transported there airborne. In the Koziol et al. (2021; *in press*) study, Cu also clustered with the natural geogenic elements, as did Mn. In the same study, Zn clustered with the likely antropogenic elements from long-range transport. However, due to its presence in ore-bearing veins in the Revvatnet surroundings, the composition of surface waters may still be impacted importantly by substratum, and not only precipitation, and this is supported by the spatial distribution of Zn concentrations in the lakes of the area (Koziol et al., 2020).

5. Summary and conclusions

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

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

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