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Water chemistry of tundra lakes in the periglacial zone of the Bellsund Fiord (Svalbard) in the summer of 2013

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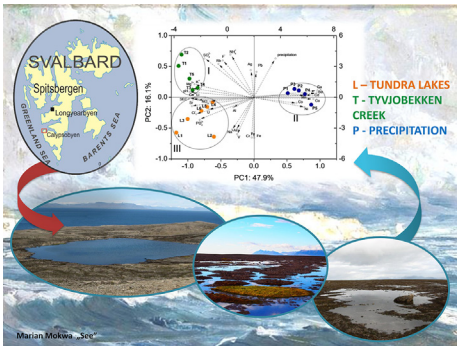
HIGHLIGHTS

- The study involved analysing tundra lakes in the Bellsund region.
- Non-sea-salt SO_4^{2-} and Mn, Co, Ni, Cu, Ga, Ba and Cd in tundra lakes are of atmospheric origin.
- Cl^- content in tundra lakes shows marine aerosol influence.
- NO_3^- present in tundra lakes may be a consequence of biological processes and permafrost thawing.

GRAPHICAL ABSTRACT



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ABSTRACT

Climate changes observed in the Arctic (e.g. permafrost degradation, glacier retreat) may have significant influence on sensitive polar wetlands. The main objectives of this paper are defining chemical features of water within six small arctic lakes located in Bellsund (Svalbard) in the area of continuous permafrost occurrence. The unique environmental conditions of the study area offer an opportunity to observe phenomena influencing water chemistry, such as: chemical weathering, permafrost thawing, marine aerosols, atmospheric deposition and biological inputs. In the water samples collected during the summer 2013, detailed tundra lake water chemistry characteristics regarding ions, trace elements, pH and specific electrolytic conductivity (SEC_{25}) analysis were determined. Moreover, water chemistry of the studied lakes was compared to the water samples from the Tyvjobekken Creek and precipitation water samples. As a final step of data analysis, Principal Component Analysis (PCA) was performed. Detailed chemical analysis allowed us to conclude what follows: (1) Ca^{2+}, Mg^{2+}, SO_4^{2-}, Sr are of geogenic origin, (2) NO_3^- present in tundra lakes and the Tyvjobekken Creek water samples (ranging from 0.31 to 1.69 mg L^{-1} and from 0.25 to 1.58 mg L^{-1} respectively) may be of mixed origin, i.e. from biological processes and permafrost thawing, (3) high contribution of non-sea-salt SO_4^{2-} > 80% in majority of studied samples indicate considerable inflow of sulphate-rich air to the study area, (4) high content of chlorides in tundra lakes (range: 25.6–32.0% meq L^{-1}) indicates marine aerosol influence, (5) PCA result shows that atmospheric transport may constitute a source of Mn, Co, Ni, Cu, Ga, Ba and Cd. However, further detailed inter-season and

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multi-seasonal study of tundra lakes in the Arctic are recommended. Especially in terms of detailed differentiation of sources influence (atmospheric transport vs. permafrost degradation).

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1. Introduction

In the face of an increase in temperature recorded in the second half of the 20th, which is anticipated to continue in the 21st century (IPCC, 2013), glacier retreat (e.g. Serreze et al., 2000; ACIA, 2005) and permafrost degradation are expected (e.g. Osterkamp and Jorgenson, 2006; Christiansen et al., 2010; Smith et al., 2010; Slater and Lawrence, 2013). The periglacial zone has become one of the most dynamically changing areas on earth (Cooper et al., 2011; López-Martínez et al., 2012; Oliva and Ruiz-Fernández, 2015).

Permafrost occurrence and active layer thickness influence hydrological processes in permafrost regions (e.g., Zhang et al., 2002; White et al., 2007; Douglas et al., 2013; Walvoord and Kurylyk, 2016). The freeze-thaw processes in aquifers determine hydrogeological conditions within catchments (Smith et al., 2005, 2007; Walvoord and Striegl, 2007; Lyon and Destouni, 2010; Cheng and Jin, 2012), whereas permafrost degradation can facilitate percolation and recharge of groundwater by atmospheric precipitation (e.g., Zhang et al., 2002; Walvoord and Striegl, 2007; Lyon and Destouni, 2010; Cheng and Jin, 2012). Thus, permafrost thawing enhances infiltration and supports deeper groundwater flow paths.

The hydrological effects of permafrost degradation in the context of changes in the surface area of lakes have been studied in Alaska (Jones et al., 2011; Karlsson et al., 2015; Arp et al., 2016) and north Siberia (Karlsson et al., 2012). The results indicated an increase in the number of thermokarst lakes and a decrease in their total surface area caused by permafrost degradation. However, Grosse et al. (2013) emphasised that thermokarst lake dynamics strongly influences the development of landscape geomorphology, hydrology, and habitats characteristic of permafrost lowlands. In the terms of chemistry, thermokarst lakes have been analysed mostly as a source of methane and carbon dioxide fluxes in summer (e.g. Walter et al., 2006; Shirokova et al., 2013; Carnevali et al., 2015). Papers discussing chemistry characteristics of thermokarst lakes (e.g. Hamilton et al., 2001; Pokrovsky et al., 2013, 2014; Manasypov et al., 2014) and the relationship between organic contaminants and metals concentrations (Manasypov et al., 2016) mostly provide results for the Canadian and Russian Arctic. Several works pertain to the Svalbard area (Lien et al., 1995; Zwoliński et al., 2008; Mazurek et al., 2012), however the number of discussed lakes and contaminants are limited, and the complex chemistry of tundra lakes in this area are still not well understood.

This paper aims at defining chemical features of water within six small arctic lakes located in Bellsund (Svalbard) (Fig. 1) in the area of continuous permafrost occurrence. The chemistry of lakes has been analysed against flowing water chemistry (Tyvjobekken Creek) and precipitation samples. Moreover, the main aim of research is to identify the sources of tundra lake water chemical composition including influence of permafrost degradation processes, atmospheric transport of contamination and marine aerosol effect. The scope of the presented study is greater compared to research available to date, as the analysis also accounts for metal contents in tundra lakes, and not only cations and anions. Moreover, an attempt is made to compare the chemistry of lake water with that of precipitation and flowing water. Since tundra lakes constitute temporary accumulation zones for compounds and part of the redepostition cascade in the sedimentary cycle of the polar oasis (Zwoliński, 2007; Zwoliński et al., 2007), it is of importance to identify their full chemical composition, particularly in the conditions of intensive changes in the periglacial environment, which are attributed to climate warming of the last half-century. Kozak et al. (2013) also notes that the studies on the concentrations of pollutants in the Arctic are a key element in the monitoring of the quality of the environment.

2. Study area

2.1. Geological and geomorphological background

The study focused on small tundra lakes in the south-western part of Wedel-Jarlsberg Land (SW Spitsbergen; Fig. 1, Fig. 2). The area is located on old Caledonian tectonic formations, renewed and transformed during the Tertiary tectonic activity (Dallmann et al., 1990; Birkenmajer, 2004). The Renardbreen block is the largest unit, composed of Protosoic slate/phyllite, weathered sandstone/conglomerate and diamictite formed of quartz, dolomite and calcareous clast (Birkenmajer, 2010). To the northern-east of Renardbreen block runs the Calypsostranda graben filled with Tertiary formations deposited over metamorphic rocks of the Kapp Lyell sequence, mainly sandstone and mudstone with interlayers of coal (Dallmann, 1988; Landvik et al., 1992; Hjelle, 1993; Birkenmajer, 2003, 2004). The main topography element of the higher parts of Wedel-Jarlsberg Land involves glacierized mid-mountain valleys of the Blomli, Scott and Renard glaciers separated by the Wijkanderberget and Bohlinryggen mountain massifs ranges reaching an elevation of 500–800 m.a.s.l. (Emil Nisonfjellet - 791 m.a.s.l.). Larger glaciers found in the area are currently in the phase of strong recession, and their forefields display considerable dynamics of morphogenetic processes. The area features ground and ablation moraines as well as

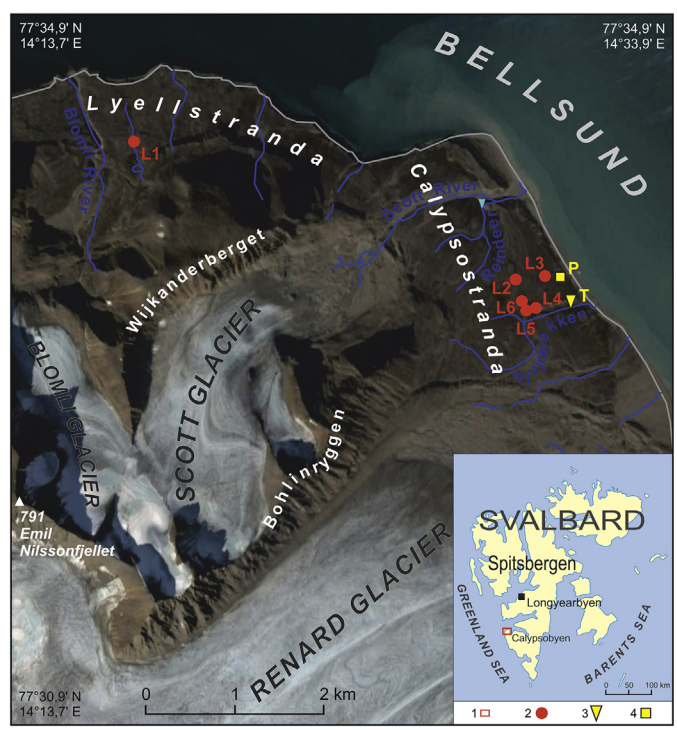

Fig. 1. Map of the studied shore of Bellsund Fiord showing the location of the studied lakes and the Tyvjobeeken Creek: 1 – location of the research area, 2 – investigated lakes (L1–L6), 3 – discharge measure and river sampling point (T), 4 – location of weather station and precipitation sampling point (P) (prepared based on http://geodata.npolar.no/#basemap-data and GoogleEarth application).

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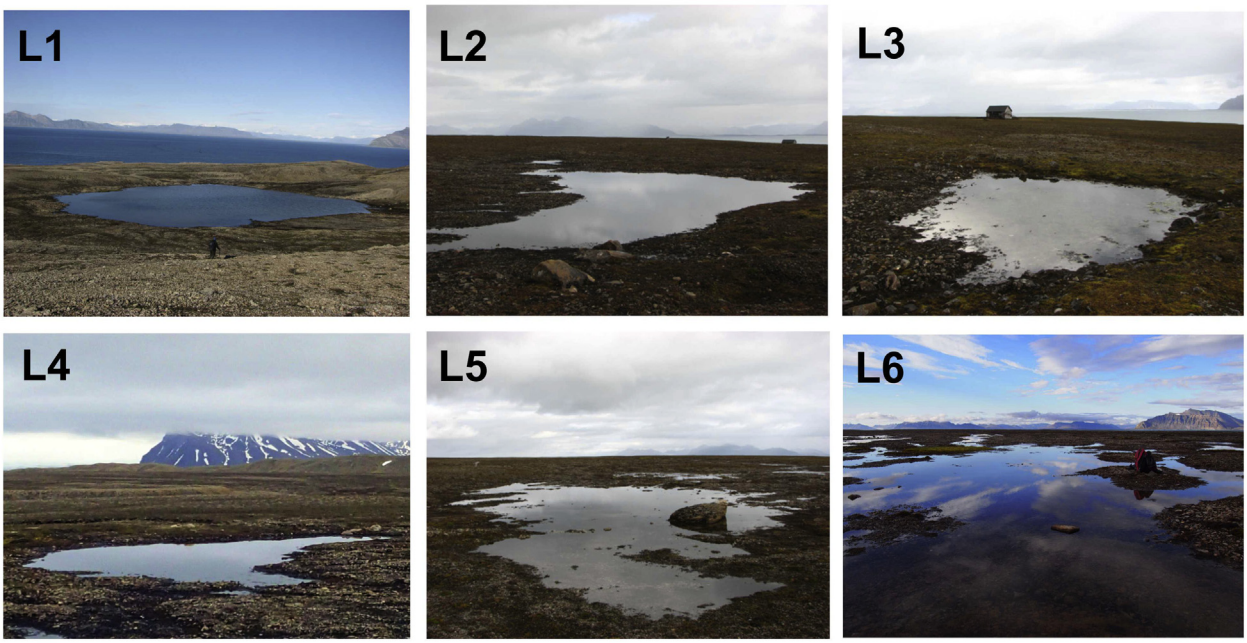

Fig. 2. Studied lakes L1–L6 at the shore of Bellsund Fiord.

inner outwash plains, which are at times accompanied by glaciofluvial crevasse fillings. These zones are remodelled by processes related to fluvioglacial erosion and deposition, as well as thermokarst phenomena. The slopes show the impact of weathering, solifluction and nival processes (Pękala, 1987; Reder, 1996; Rodzik et al., 2013). The northern and north-eastern part of the study area features raised isostatic sea plains reaching up to 2 km into the land. They are composed of a system of marine terraces that are diverse in terms of age and hypsometry, and whose surfaces are shaped by solifluction and ice segregation (Szczęsny et al., 1989; Landvik et al., 1992).

2.2. Meteorological and hydrological background

In the Bellsund region cyclonic circulation (51%) prevails over the anticyclonic (47%). The most frequently occurring circulation is from sector NW, N and NE (Gluza and Siwek, 2012). These conditions are locally subject to transformation owing to the combined impact of both marine and terrestrial environment, which features considerably diverse lay of the land. Mean air temperature in the NW part of Wedel Jarlsberg Land in the summer seasons of 1986–2006 (estimated against 15 seasons) was at 5.1 °C (Gluza and Siwek, 2013). Mean precipitation

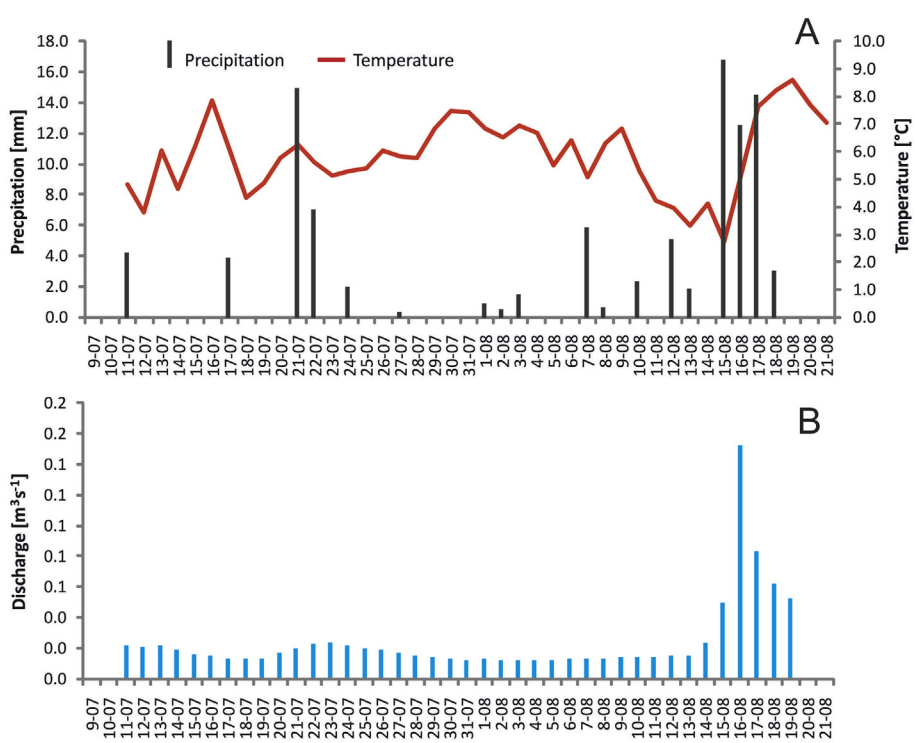

Fig. 3. Weather conditions (A) and discharges (B) of Tyvjobekken Creek in the period from July 12th, to August 23rd, 2013.

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for the ablation seasons in the period of 1986–2006 amounted to 34.6 mm. The highest was recorded in 1994, 82.3 mm, whereas the lowest in 1998–10.0 mm. Average wind velocity in the summer seasons of the study period amounted to 4.3 $m \cdot s^{-1}$. The highest value was recorded in 2005, 6.7 $m \cdot s^{-1}$, and the lowest in 1995–2.7 $m \cdot s^{-1}$ (Gluza and Siwek, 2013).

All of the study lakes are located at an elevation of 25–80 m.a.s.l. and within the reach of non-glacierized drainage basins developed in raised marine terraces (Lyellstranda – lake L1 Calypsostranda – lakes L2–L6) (Fig. 1). Lakes L4, L5 and L6 are situated in close proximity to one another at an elevation of 30 m.a.s.l, 600–700 m away from the water of Recherche Fjord, in the western part of the mid-section of the Tyvjobekken Creek drainage basin (Fig. 1). The river is 1.2 km long and its drainage basin covers an area of 1.3 km^2. Its course begins with periodic streams emerging at the foot of the Bohlinryggen mountain massif range, at an elevation of 100 m.a.s.l., and the river debouches into Recherche Fjord (Bartoszewski, 1998; Kociuba, 2015). The Tyvjobekken Creek is characterised by a complex snow-permafrost-rain alimentation system.

Reservoirs found within the drainage basin of the Tyvjobekken Creek are endorheic, shallow (maximum depth – 25 cm) and in summer tend to dynamically shift their surface area, at times disappearing completely. The largest one is lake L6 (Fig. 2). Its surface area is difficult to estimate owing to its considerably variable shoreline and large number of islands, however, during wet weather periods it may exceed 9000 m^2. Reservoir L2 has small surface area and depth, and features considerably developed shoreline. Object L3 (Fig. 2) constitutes the smallest of the analysed basins and the nearest one from the Fjord water (350 m). Its surface area amounts to 32 m^2, and its maximum depth reaches 30 cm. Lake L1 is the highest of all studied lakes, located within a marine terrace at an elevation of 70–80 m a.s.l. (Lyellstranda), 900 m away from the Recherche Fjord (Fig. 1, Fig. 2). The surface area of the lake amounts to 7860 m^2 and it is up to 45 cm deep. The basin is stable and periodically drained at the beginning of summer. The analysed lakes, under observation in the summer seasons of 2013–2016, showed considerable seasonal surface area fluctuation. The smallest ones (L2, L4 and L5) would periodically disappear.

All of the analysed lakes are located within a periglacial zone, at a distance of approx. 2 km from the nearest glaciers. These shallow basins lie within the active permafrost layer, depth of which (studied in 1986–2012) ranged from 40 to 225 cm (Repelewska-Pękalowa et al., 2013). In recent years thickness of the active permafrost layer exceeded 150 cm on a regular basis.

3. Materials and methods

3.1. Sampling design

Surface water samples were collected from 6 lakes on August 21st, 2013 (L1–L6 in the Figs. 1, 2, 4, 5 and 6). In order to compare lake water chemistry with the chemical composition of flowing water and precipitation, samples were taken from the Tyvjobekken Creek and at

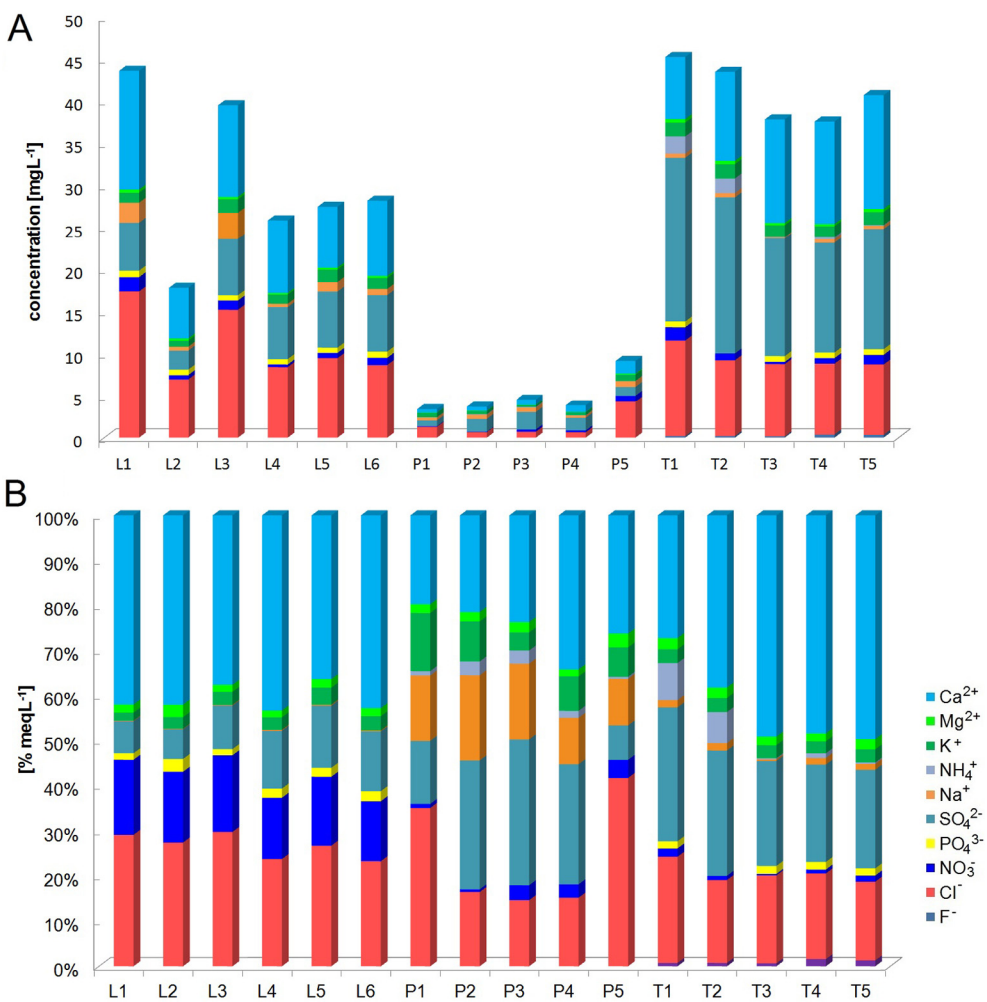

Fig. 4. Distribution of determined ions in the studied water samples: L - tundra lakes; P - precipitation, T - Tyvjobekken Creek. A. Ion concentrations in mg L^{-1} B. Percentage of ion concentrations in meq L^{-1}.

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Table 1
Non-sea salt (nss) SO_4^{2-} concentrations (meq L^{-1}) in studied water samples of the studied shore of Bellsund Fiord and nssSO_4^{2-} percentage of total sulphate concentration.

	L1	L2	L3	L4	L5	L6	P1	P2	P3	P4	P5	T1	T2	T3	T4	T5
nssSO_4^{2-}	0.069	0.027	0.095	0.104	0.111	0.114	0.011	0.030	0.042	0.030	0.010	0.372	0.360	0.266	0.247	0.273
% of total SO_4^{2-}	57.7	57.1	68.3	80.9	80.0	81.9	74.1	94.1	95.3	94.1	43.6	91.8	93.2	91.4	90.9	91.8

the Hellmann rain gauge with inlet ring surface of 200 cm^2, between August 17st and 21st, 2013 (T1–T5 and P1–P5 in the Figs. 4, 5 and 6). Total number of samples is $n = 16$ and include: 6 lake samples (6 L), 5 precipitation samples (from 14 to 288 mL per sample) and 5 flowing water samples (Tyvjobekken Creek) (5 L). Samples were taken with special care to avoid contamination (e.g. by using the polyethylene gloves). Samplers were rinsed with river or lake water three times and then filled without air bubbles to prevent the loss of analytes to headspace. Samples were collected manually just below the water surface. All collected material was stored and transported to the laboratory in Poland in airtight samplers and stored in low temperatures (approx. +4 °C). In the reference to the Good Laboratory Practice the study involved analysing blank samples (deionised water stored in the same containers as field samples). This element of analytical procedure is to verify impact of sample container on chemical composition of water samples (e.g. adsorption of analytes on the container walls).

The collection of samples was preceded by meteorological measurements performer with the automatic weather station (Campbell Scientific CR10 datalogger), which recorded data every 10 min. The weather station was located within the raised marine terrace near the Calypso settlement, approximately 200 m from the seashore at an altitude of 23 m a.s.l. (Fig. 1). Runoff from the Tyvjobekken creek was established against on a rating curve based on periodical discharge measurements and the corresponding water levels. Changes in water levels were estimated based on measurements obtained with a water level logger equipped with a pressure sensor (Schlumberger Water Services) set to 10-minute intervals. Water flow velocity was measured twice a week with a rotor current meter Hega II and an acoustic digital current meter OTT ADC.

3.2. Laboratory methods

The concentrations of metals and ions in water samples were determined with inductively coupled plasma mass spectrometry Thermo XSERIES 2 ICP-MS (Thermo Fischer Scientific, Bremen, Germany) and DIONEX 3000 chromatograph (DIONEX, USA), respectively. Physicochemical parameters such as pH and specific electrolytic conductivity (SEC_{25}) were determined by the electrochemical method with the application of a microcomputer pH/conductometer CPC-411 by Elmetron, electrode type EPS-1, and conductivity sensor EC 60. Accuracy of the pH microcomputer/Elmetron CPC-411 conductometer was ±0.01 for pH and ±0.25% of conductivity value [μS cm^{-1}] for conductivity. Technical specifications, as well as basic validation parameters of the analytical procedures for metal and ions are summarized in Supplementary Material 1.

The contribution of sea-salt and non-sea-salt (nss) component of SO_4^{2-} was calculated in the following way: nss$SO_4^{2-} = SO_4^{2-} - (SO_4^{2-} / Cl^-)_{seawater} * Cl^-$. The $(SO_4^{2-} / Cl^-)_{seawater}$ ratio is the equivalent ratio of SO_4^{2-} to Cl^- in global mean seawater, which is 0.103 (Stumm and Morgan, 1996). Calculations were based on the assumption that all deposited chlorides are of marine origin, following Krawczyk et al. (2008), Chmiel et al. (2011), Stachnik et al. (2016), Nawrot et al. (2016).

3.3. Statistical analysis

Pearson's correlation coefficients (r) were calculated to detect pairwise relationships among metals, ions, pH and SEC_{25} in the investigated water samples. Correlation coefficients was considered statistically significant at $p < 0.05$. Principal Component Analysis (PCA), based on the correlation matrix was performed to identify patterns across all physicochemical data. PCA and Pearson's correlation coefficients were computed with the software package STATISTICA 7 (StatSoft Inc.,) and Excel 2010 (Microsoft), respectively.

4. Results

4.1. Weather and hydrological conditions at the time of sampling

Compared to average weather conditions in summer months (observed from 1986, Franczak et al., 2016), the ablation season of 2013 (measurements taken from Jul 12th to Aug 23rd, 2013, Fig. 3A) was warmer and less wet. Mean air temperature in this period amounted 5.9 °C, with the maximum daily temperature of 8.6 °C recorded on August 17th, and the minimum of 2.7 °C on August 13th. Total

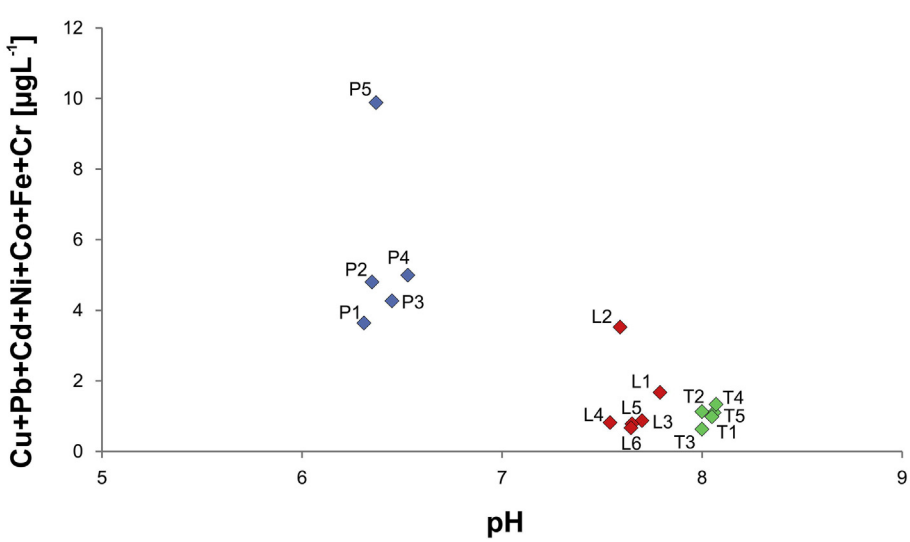

Fig. 5. Ficlin Diagram for studied water samples (Naseem et al., 2014).

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Table 2
Values of Pearson's correlation coefficient (r) regarding total concentrations of the investigated elements (selected ions, metals, pH and SEC). Very strong correlations ($0.8 < |r| \leq 1$), important in significance at $p < 0.05$, are in bold.

	pH	SEC	F^-	Cl^-	NO_3^-	PO_4^{3-}	SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Li	Al	V
pH	1.00														
SEC	**0.97**	1.00													
F^-	0.60	0.50	1.00												
Cl^-	0.80	**0.90**	0.20	1.00											
NO_3^-	0.61	0.69	0.28	**0.81**	1.00										
PO_4^{3-}	**0.81**	**0.84**	0.28	0.77	0.55	1.00									
SO_4^{2-}	**0.81**	0.74	0.79	0.50	0.51	0.41	1.00								
Na^+	0.19	0.35	−0.30	0.69	0.54	0.31	−0.11	1.00							
NH_4^+	0.37	0.30	0.41	0.20	0.41	−0.07	0.73	−0.16	1.00						
K^+	**0.90**	**0.92**	0.53	**0.81**	0.66	0.66	**0.82**	0.31	0.45	1.00					
Mg^{2+}	**0.93**	**0.93**	0.57	**0.84**	0.80	0.68	**0.83**	0.28	0.53	**0.90**	1.00				
Ca^{2+}	**0.92**	**0.96**	0.55	**0.85**	0.63	0.79	0.68	0.35	0.14	**0.84**	**0.87**	1.00			
Li	**0.83**	0.80	0.30	0.69	0.49	0.59	0.63	0.18	0.47	0.72	**0.83**	0.69	1.00		
Al	0.35	0.32	0.22	0.17	0.18	0.45	0.15	−0.08	−0.17	0.29	0.24	0.35	0.20	1.00	
V	0.15	0.28	−0.29	0.51	0.46	0.31	−0.16	0.48	−0.14	0.02	0.29	0.31	0.29	−0.12	1.00
Cr	0.02	0.04	−0.26	0.17	0.08	0.18	−0.29	0.16	−0.24	−0.15	0.09	0.03	0.37	0.02	0.59
Mn	−0.68	−0.71	−0.31	−0.61	−0.37	−0.62	−0.49	−0.23	−0.19	−0.61	−0.63	−0.65	−0.63	−0.22	−0.15
Fe	−0.02	−0.03	−0.30	0.06	0.04	0.16	−0.35	0.02	−0.23	−0.22	0.03	−0.07	0.36	0.12	0.53
Co	−0.60	−0.54	−0.28	−0.36	−0.13	−0.52	−0.44	−0.07	−0.18	−0.40	−0.43	−0.50	−0.48	−0.10	0.02
Ni	−0.73	−0.70	−0.34	−0.51	−0.24	−0.63	−0.59	−0.13	−0.21	−0.63	−0.56	−0.65	−0.51	−0.16	0.08
Cu	**−0.86**	**−0.84**	−0.37	−0.67	−0.37	−0.76	−0.63	−0.21	−0.22	−0.74	−0.71	−0.79	−0.70	−0.28	−0.08
Ga	**−0.93**	**−0.95**	−0.39	**−0.82**	−0.53	**−0.85**	−0.64	−0.28	−0.23	**−0.87**	**−0.85**	**−0.87**	**−0.84**	−0.37	−0.22
As	0.21	0.18	−0.29	0.27	0.17	0.30	−0.11	0.32	0.00	0.15	0.19	0.04	0.54	0.26	0.11
Rb	0.57	0.50	0.47	0.36	0.40	0.16	**0.82**	−0.17	**0.84**	0.61	0.70	0.40	0.69	−0.08	0.01
Sr	**0.90**	**0.93**	0.27	**0.86**	0.55	0.77	0.60	0.41	0.23	**0.86**	**0.83**	**0.88**	**0.85**	0.37	0.24
Ag	0.08	0.07	0.41	−0.06	0.16	−0.30	0.31	−0.18	0.29	0.16	0.29	0.15	0.15	−0.03	0.02
Cd	**−0.83**	**−0.81**	−0.30	−0.67	−0.36	−0.75	−0.58	−0.22	−0.24	−0.69	−0.70	−0.74	−0.75	−0.24	−0.14
Ba	**−0.91**	**−0.91**	−0.38	−0.76	−0.46	**−0.83**	−0.63	−0.25	−0.23	−0.80	−0.79	**−0.84**	**−0.81**	−0.32	−0.18
Pb	0.04	−0.01	0.22	−0.05	0.15	−0.34	0.34	−0.21	0.55	0.17	0.24	−0.04	0.28	−0.04	−0.10
U	0.59	0.69	0.11	**0.80**	0.69	0.46	0.39	0.52	0.26	0.51	0.70	0.71	0.60	−0.03	0.78

precipitation amounted to 98.9 mm. There were 26 days with rain (43%) and the highest daily precipitation of 16.8 mm was recorded on August 13th. Furthermore, a sequence of four days (13th to 16th of August) made up almost half (47%) of total precipitation in the study period. Mean daily wind velocity amounted to 3.7 ms^{-1}, with the highest recorded on August 9th – 8.2 ms^{-1}, and the lowest on August 23rd – 0.6 ms^{-1}. Westerly wind prevailed over the course of the entire study period, with slightly lower frequency of easterly and south-westerly wind.

Mean discharge in the Tyvjobekken Creek throughout the study season of 2013 (Jul 12th – Aug 23rd, 2013) amounted to 0.025 $m^3 s^{-1}$, total runoff – 85 thousand m^3 and runoff layer – 65 mm. High water level occurred in July (15th and 23rd, discharges – 0.022 $m^3 s^{-1}$ and 0.023 $m^3 s^{-1}$ respectively) and August (maximum of 0.084 $m^3 s^{-1}$ on August 17th) (Fig. 3B). The sampling period (Aug 13th–21st) coincided with the high water levels of August. The Tyvjobekken Creek is a periglacial river and its maximum discharges are largely determined by the spring season. However, at times a considerable rise in discharge may also occur during polar summer and autumn in consequence of increased precipitation and permafrost melting (Bartoszewski, 1998). Over the series of ablation seasons under analysis, duration of which varied, mean discharges in the Tyvjobekken Creek ranged from 0.084 $m^3 s^{-1}$ (1986) to 0.014 $m^3 s^{-1}$ (1987).

4.2. Basic inorganic analysis

The values of pH range from 7.54 to 7.79; 6.31–6.53, and 8.00–8.07 for tundra lakes, precipitation and the Tyvjobekken Creek respectively (Supplementary Material 2). In turn, values of SEC_{25} range from 184 to 321 μS cm^{-1}, 7.40–37.3 μS cm^{-1} and 270–287 μS cm^{-1} for tundra lakes, precipitation and the Tyvjobekken Creek respectively. Ion concentrations and their percentage in each sample are presented in Fig. 4. All of selected inorganic analysis results are also listed in Supplementary Material 2. Tundra lakes L1 and L3 have the highest concentration of ion sums but still remain within the low mineralisation range (<100 mg L^{-1}). Lakes L4–L6 represent a group of lakes, where sum of ion ranges from 34.0 to 36.0 mg L^{-1} and is similar to ion concentration recorded in the samples taken from the Tyvjobekken Creek (37.6–45.3 mg L^{-1}) (Fig. 4A). The following ions were found to have the highest percentage share in the samples: Ca^{2+}, Cl^- and SO_4^{2-}. However, percentage distribution of selected ions [% in meq L^{-1}] differ in tundra lakes, precipitation and the Tyvjobekken Creek. Percentage of Ca^{2+} was in the range of 40.2–47.6%; 19.7–34.2% and 27.2–49.6% for tundra lakes, precipitation and the Tyvjobekken Creek water samples respectively. In the case of Cl^- percentage content, the value ranges are as follows: 25.6–32.0%; 14.7–41.8%; 17.5–23.5% for tundra lakes, precipitation and Tyvjobekken Creek water samples respectively. Moreover, percentage of SO_4^{2-} was found to be significant in precipitation and the Tyvjobekken Creek water samples and ranged from 7.62% to 32.3%, and from 21.6 to 29.7% respectively (Fig. 4B).

Since the analysed lakes are located in the area of permafrost occurrence, it is also worth to draw attention to NO_3^- contents (Walvoord and Striegl, 2007). As far as NO_3^- concentration in tundra lakes is concerned, it ranged from 0.31 to 1.69 mg L^{-1},whereas in the Tyvjobekken Creek it was in the range of 0.25–1.58 mg L^{-1} (Fig. 4A).

Moreover, our results indicate very high percentage of $nssSO_4^{2-}$ contribution: 57.7–81.9% in tundra lakes, 43.6–94.1 in precipitation samples and 90.9–91.8% in the Tyvjobekken Creek (Table 1).

4.3. Trace metal analysis

Total concentration of metals in water samples ranged from 52.50 to 73.5 μg L^{-1} in lakes and the Tyvjobekken Creek (Supplementary Material 3). However, their concentration in precipitation samples was higher and ranged from 68.0 to 147 μg L^{-1}. One should note that higher metal concentration coincided with lower pH, ranging from 6.31 to 6.53 in precipitation, 7.54–7.79 in lakes and 8.00–8.07 in the Tyvjobeeken Creek (Fig. 5). The level of occurrence of metals in water shows prevalence of Sr and Ba in all of the analysed samples, and Cu, Ga and Mn in precipitation samples. Sr in the lakes and the creek ranged from 44.2 to 69.1 μg L^{-1}, whereas Ba from 1.43 to 6.48 μg L^{-1}. In precipitation

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Cr	Mn	Fe	Co	Ni	Cu	Ga	As	Rb	Sr	Ag	Cd	Ba	Pb	U
1.00														
−0.06	1.00													
0.94	−0.05	1.00												
0.27	0.55	0.19	1.00											
0.39	0.63	0.35	**0.93**	1.00										
0.17	0.73	0.15	**0.89**	**0.96**	1.00									
−0.10	0.77	−0.10	0.60	0.73	**0.89**	1.00								
0.60	−0.21	0.66	−0.13	−0.04	−0.19	−0.33	1.00							
0.06	−0.32	−0.03	−0.18	−0.27	−0.35	−0.44	0.08	1.00						
0.10	−0.68	0.05	−0.54	−0.69	**−0.84**	**−0.94**	0.37	0.43	1.00					
0.01	0.01	0.01	0.18	0.16	0.12	0.03	−0.20	0.35	−0.01	1.00				
0.07	0.77	0.03	**0.89**	**0.93**	**0.99**	**0.89**	−0.28	−0.36	**−0.83**	0.14	1.00			
−0.01	0.75	−0.03	0.77	**0.85**	**0.96**	**0.97**	−0.31	−0.39	**−0.91**	0.07	**0.97**	1.00		
0.23	0.15	0.14	0.52	0.45	0.35	0.10	0.10	0.62	0.02	0.57	0.34	0.24	1.00	
0.20	−0.44	0.13	−0.33	−0.36	−0.51	−0.58	0.00	0.37	0.64	0.16	−0.53	−0.57	0.00	1.00

samples the relation between these two metals is different and Sr ranged from 2.60 to 7.68 μg L^{-1}, and Ba from 59.4 to 125 μg L^{-1}.

Correlation matrix of particular metals can be found in Table 2. Very strong correlations ($0.8 < |r| \leq 1$) have been observed for Ba and the following metals: Cu (0.96), Ga (0.97), Ni (0.85), Cd (0.97) (Table 2). Therefore, these metals correlate strongly with one another, and negatively with Sr, pH, SEC as well as with almost all cations and anions. Of all the analysed samples, the highest concentration levels of Ba, Cu, Ga, and Cd, have been observed in the precipitation samples. In the second group of metals, associated with lakes and creek waters, very strong correlations have been observed between: Sr and Li (0.85), and Fe and Cr (0.94). However, only the first pair correlates strongly with pH, SEC and Cl^-, K^+, Mg^{2+}, Ca^{2+}.

4.4. Principal component analysis

The PCA has been created for the entire data set. All 31 hydrogochemical variables have been accounted for in the analysis. Variables taken into consideration include major ions, metals, pH, SEC and precipitation amount. The selected two principal components represent >60% of the variance. PC1 is strongly positively correlated with the following variables: Mn, Co, Ni, Cu, Ga, Ba and strongly negatively correlated with K^+, Mg^{2+}, Ca^{2+}, PO_4^{3-},SO_4^{2-}, Cl^-, Li, Sr, pH and SEC. PC2 is positively correlated with NH_4^+, F^-, SO_4^{2-}, Rb, Ag, Pb and precipitation, and negatively correlated with Na^+, NO_3^-, As, V, Cr and Fe (Fig. 6). Based on the projection of each data point on the plane of the two principal components (Fig. 6), the water samples can be divided into three groups, represented by tundra lake water, creek water and precipitation water. On the PCA biplot (Fig. 6) we can clearly distinguish three groups of samples, i.e. lake, precipitation and flowing water samples. These groups did not show visible correlation with PC1, excluding all precipitation samples. Precipitation samples are strongly positively correlated with PC1 and at the same time with the following variables: Mn, Co, Ni, Cu, Ga, Ba and Cd.

5. Discussion

5.1. Marine aerosols and atmospheric transport influence

Location of the lakes (200 m–900 m from the sea-shore) allows us to draw a hypothesis that the studied tundra lakes are under the marine aerosol influence. The lowest concentration in L2 may be related to its small surface and depth. As concluded in the authors' study results concerning lakes in Mongolia (Szopińska et al., 2016; Szumińska, 2016), stability of a lake may constitute one of major factors determining its chemical composition. In drying lakes, bed sediments can be deflated along with compounds that form evaporites during desiccation, particularly under the influence of intense wind of the Arctic tundra. de Caritat et al. (2005) emphasises that the lack of vegetation in the Arctic may cause important fluxes of mineral dust (together with its chemical elements content) to the atmosphere.

Because of the non-flowing status of the studied tundra lakes they are adapted to accumulate compounds transported via dry and wet deposition. Tundra lakes, as the entire Arctic tundra, constitute the alimentation area of marine aerosols (Zwoliński et al., 2008). The accumulated compounds may be redeposited both as a result of solution circulation during the active layer thawing and through transport along thermal gradients within permafrost (Kokelj and Burn, 2003; Kokelj et al., 2005). While not all chlorides identified in lake waters found in the area are of maritime origin (Krawczyk et al., 2008; Mazurek et al., 2012), the contribution of these Cl^- is prevailing and exceeds 80% (Mazurek et al., 2012). The concentrations of Cl^- in the analysed lakes were found to be at a similar level to the results obtained for the lakes in the Svalbard area, as presented by the above-mentioned authors. Lakes L1–L6, on the other hand, showed lower concentrations of Na^+ (Supplementary Material 2) than in the lakes investigated by Krawczyk et al. (2008) and Mazurek et al. (2012), which are, nevertheless, comparable to the results obtained by Manasypov et al. (2014). Manasypov et al. (2014) also points out that, compared to continental (southern) thermokarst basins, lakes in the Arctic coastal continuous permafrost zone are enriched in alkali metals (Na^+, K^+). As in the

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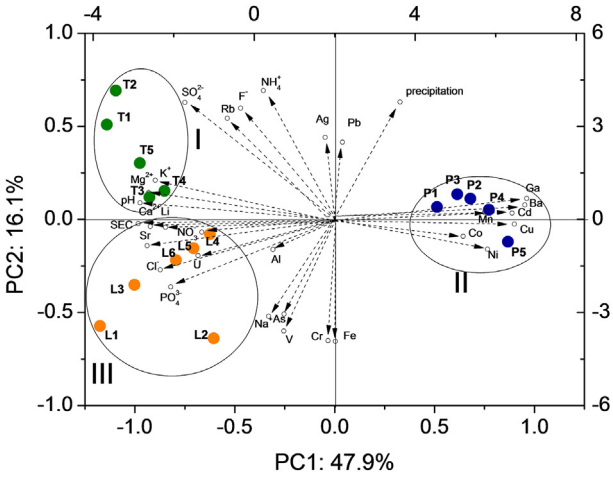

Fig. 6. PCA biplots for various data sets. Projection of environmental variables (bottom X axis and left Y axis) and cases - sampling points (upper X axis and right Y axis) on the plane of two principal components.

case of Cl^-, lakes L1 and L3 were found to have the highest Na^+ concentration (Fig. 4, Supplementary Material 2) and are comparable to the results obtained by Manasypov et al. (2014) in lakes located within the shoreline zone (2.62 mg L^{-1}). The former lake (L3) is located approx. 900 m away from the fiord and clearly stands out due to its large surface area (7860 m^2) and stability, which translates into a consistent process of enrichment in ions related to the impact of marine aerosols. It is also worth noting that Na^+ and K^+ both constitute a considerable share (approx. 20–30%) in total sum of anions and cations recorded in precipitation water (Fig. 4B). Despite generally low content of anions and cation in precipitation samples, their composition allows us to draw a conclusion that Na^+ and K^+ found in the analysed lakes are of atmospheric origin. Nawrot et al. (2016) also points out that strong correlation between Cl^- and Na^+ in snow of the Hans glacier is indicative of the marine source of these two ions, and the same can be observed in the case of water samples from Bellsund (Table 2).

Atmospheric transport of contaminants has been thus far studied in the context of the anthropogenic impact on water chemistry in the area of Svalbard, owing to scarcity of local emission sources (e.g. Krawczyk et al., 2008; Kozak et al., 2015; Kosek and Polkowska, 2016; Lehmann et al., 2016; Nawrot et al., 2016). Elements found in arctic precipitation are frequently of both natural and anthropogenic origin, and both these sources may contribute to the wet (soluble) and dry (aerosols, dust) components of precipitation (de Caritat et al., 2005). As illustrated with Fig. 6, the precipitation samples are positively correlated with PC1 and at the same time show positive correlation with Mn, Co, Cu, Cd, Ni, Ga and Ba. All the listed contaminants were found to be at much greater concentration in precipitation water than in lakes and the Tyvjobekken Creek (Supplementary Material 3). This occurrence can be associated with the sea-spray influence on precipitation rather than long range atmospheric transport of these metals to Spitsbergen, which was also mentioned by Kozak et al. (2015). This means the contaminants may be products of fuel combustion on ships (Zhan et al., 2014). Moreover, Pacyna (1995) argues metal industry to be the main source of Cd, Cu, Ni, As, in the Arctic. Nawrot et al. (2016) mentioned, that S in the form of SO_4^{2-} is the predominant ion present in Arctic Haze. High contribution of nssSO_4^{2-} >80% in most of studied samples (Table 1) is indicative of a considerable inflow of sulphate-rich air to the study area. The origin of this occurrence may be of mixed nature. Nawrot et al. (2016) argues that the primary source of SO_4^{2-} around Hornsund Fiord lies with agricultural fires in Eastern Europe. Krawczyk et al. (2008), on the other hand, demonstrates that the influx of contaminated air over to the western shore of Spitsbergen comes from the Greenland Sea and the Norwegian Sea, with ships being the main emitters. Nevertheless, clear differentiation between anthropogenic input and chemical denudation of sulfates constitutes a task for the further research.

5.2. Geochemical and permafrost factors

The Arctic area is known to be highly susceptible to mechanical and chemical weathering of outcropping rocks, soil dust deflation in snow-free areas (de Caritat et al., 2005), as well as washing out during increased discharges related to ablation season (Zwoliński et al., 2008; Rachlewicz et al., 2016). Deflated and washed out material constitutes yet another natural source of chemical elements in water, although the supply of the said material can be preceded by its accumulation on snow and glaciers (Hinkley, 1994; Nawrot et al., 2016).

The analysed lakes are located within a non-glacierised zone at the forefield of the Scott and Renard glaciers. Here geochemical conditions are largely affected by moraine and sandur material washed out from the forefields of glaciers, as well as by bed rocks that form the raised marine terraces, on which the lakes are located. These are mainly sedimentary rocks related to marine deposition that may constitute a source of carbonates and carbon. While the analysis of carbonates was not part of the study scope, high Ca^{2+} content in water samples from lakes and the Tyvjobbeken Creek, compared to precipitation water, appears to be indicative of its geogenic origin. The PCA analysis, on the other hand, indicated a strong negative correlation between Ca^{2+} and a group of metals that have strong positive correlation with PC1, which determines the composition of precipitation water (Fig. 6). Furthermore, Ca^{2+} in the analysed samples shows very strong correlation with pH, SEC, K^+ and Mg^{2+} and the following metals: Sr, U and Li (Table 2). These elements occur both in the analysed lakes and the Tyvjobekken Creek, but are scarce in precipitation water. The geogenic origin of Ca^{2+} found in lake waters in the Billefjorden area is indicated by Mazurek et al. (2012), who argues it comes from the dissolution of calcium carbonate ($CaCO_3$). The high content of Ca^{2+} and Mg^{2+} in ground waters in the Bellsund region has been also pointed out by Chmiel et al. (2013), as the result of carbonate mineral dissolution. Research conducted by Mazurek et al. (2012) show that the lakes in the Bille Fiord, similarly to the lakes analysed in the Bellsund Fiord L1–L6, show the prevalence of Ca^{2+}, whose presence in water (including Mg^{2+}) must be balanced by SO_4^{2-}. Sulphate in the analysed lakes L1-L2, similarly to the results presented by Mazurek et al. (2012), is one of the most important anions in lake water (Fig. 5), which shows strong correlation with Ca^{2+} and very strong with Mg^{2+} (Table 2). Sr is another important component of geogenic origin occurring in much higher concentration in lakes L1–L6 and the Tyvjobekken Creek than in precipitation water (Supplementary Material 3). This is evident from strong correlation between the occurrence of Sr and ions Ca^{2+}, Mg^{2+} and SO_4^{2-} of geogenic origin. The occurrence and correlation of Ca^{2+} and SO_4^{2}, as well as their positive correlation with SEC (Table 2) were indicated by Mazurek et al. (2012) as factors suggesting the geogenic origin of sulfates. The enrichment of thermokarst lake waters in the Arctic seashore in alkaline-earth (Ca^{2+}, Sr) is also indicated by Manasypov et al. (2014), and its origin from mineral dust is emphasised by de Caritat et al. (2005).

Wind and water erosion in the analysed area is determined by the occurrence of permafrost and its active layer thawing depth. At present, as the nearby glaciers are retreating and the source areas of streams have been cut off from the direct supply of thawing water, the main sources of stream recharge, as well as surface and subsurface run off are connected with permafrost thawing, snowmelt and wet precipitation. According to Bartoszewski et al. (2013) these are three main feeding sources of the Tyvjobbeken Creek during high water levels in spring. Taking into consideration current hydrometeorological conditions, it can be assumed that in summer the contribution of permafrost in the feeding of surface waters is largest (Bartoszewski, 1998), and its infiltration limiting influence smallest. Studies conducted within

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Calypsostranda indicate that thawing depth of the active layer in the study area is considerable and exceeds 150 cm (Repelewska-Pękalowa et al., 2013). However, it should be emphasised that detailed studies carried out in selected areas of the Arctic indicate considerable variability in terms of both permafrost occurrence and thawing depth of its active layer, as well as its dependency of local factors, such as: topography, lithology, geothermal heat flow, vegetation cover, distance to ocean, wind and snow cover (Humlum et al., 2003; Dobiński and Leszkiewicz, 2010; Kasprzak et al., 2016; Sobota et al., 2016).

The presence of permafrost in the bed of analysed lakes is evident from their morphological features, i.e. considerably variable shoreline, small depth and marked surface area fluctuation (particularly in the case of L2–L6). Variability of shoreline is connected with the presence of patterned ground (Fig. 6), considered characteristic of permafrost occurrence areas. As far as surface area fluctuation is concerned, it is not only a result of shifts in the volume of snowmelt and wet precipitation, but also the thawing depth of permafrost.

The result point to (Fig. 4) a similar concentration of NO_3^- level in tundra lakes and in the Creek Tyvjobekken. In precipitation samples the level of NO_3^- is approximately ten times lower than in the mentioned samples. N in this form can be of mixed origin, i.e. from biological processes and permafrost seasonal thawing. Wynn et al. (2007) studied the influence of microbiological activity on NO_3^- concentration in water samples from glacierised areas. His results indicate that considerable share of NO_3^- found in subglacial meltwater comes, first and foremost, from microbiological nitrification of NH_4^+ and mineralisation of organic nitrogen in layers of snow. However, the authors also emphasise that in late summer, as the supply of meltwater is more limited, concentration of nitrates in meltwater tends to increase, which suggest that there is most likely an additional source feeding nitrates (aside from microbiological activity) to subglacial water. Chmiel et al. (2013) on the other hand, demonstrates that the high concentrations of P and N in the vicinity of lakes in the Bellsund region can be associated with a bird colonies and reindeer herds. Although nitrogen and carbon amount in watersheds affected by the permafrost active layer thawing have been described mostly in relation to in-land permafrost regions (e.g., Carey, 2003; O'Donnell and Jones, 2006; Petrone et al., 2006; Frey et al., 2007; Frey and McClelland, 2009; Keller et al., 2010; Bagard et al., 2011; Douglas et al., 2013; Larouche et al., 2015), we can assume that nitrogen of biogenic origin could be stored in the active layer. Hence, permafrost degradation processes in summer may constitute a secondary source of nitrogen compounds in the studied tundra lakes.

6. Conclusions

This work presents the chemical inorganic analysis of the tundra lakes located at the shore of Bellsund. The study showed a considerable contribution of elements of geogenic origin: Ca^{2+}, Mg^{2+}, SO_4^{2-}, Sr, which is in line with the findings of other authors, who have indicated that these elements tend to prevail in the tundra lakes of Svalbard. Moreover, the analysis of metals concentrations revealed the following group of elements: Mn, Co, Cu, Cd, Ni, Ga and Ba, all of which were found to be of atmospheric origin. In this group, Co, Cu, Cd and Ni, which represent heavy metals, are considered to be particularly threatening to the environment. Identifying their source, be it of short-distance or long-distance atmospheric transport, requires further research, particularly in relation to dry atmospheric deposition. Chemical composition of tundra lakes has also been found to be considerably affected by marine aerosols, which enrich their water in chlorides. The study has also revealed that tundra lakes may potentially constitute deposition areas for nitrogen released, among others, in biological processes and permafrost thawing.

Factors affecting the status of lake water chemistry recommended for further investigation include detailed distinction of permafrost influence paired with the atmospheric transport of compounds. As non-glacierised areas of Svalbard expand and permafrost degradation proceeds, we may expect to see an increase in the importance of tundra lakes and their impact on migration of compounds. Taking into consideration the anticipated depth increase of the active layer in the 21st century, the number of tundra lakes are likely to increase. These lakes will act as geochemical traps for wet and dry deposition compounds, as well as compounds of geogenic and biogenic origin and those released from permafrost. Moreover, the said lakes will constitute places of secondary emission of the accumulated compounds into water and atmosphere.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2017.10.045.

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