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# Comparison of hydrochemistry and organic compound transport in two non-glaciated High Arctic catchments with a permafrost regime (Bellsund Fjord, Spitsbergen)

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## ABSTRACT

An increase in air temperature related to climate change results in the retreat of glaciers, the degradation of permafrost, and the expansion of glacier-free areas in the polar regions. All these processes lead to changes in the Arctic landscape. They influence the hydrochemistry of streams and rivers fed by glaciers and thawing permafrost. In this study, we examine eighty two water samples from two non-glaciated catchments with snow-permafrost regime: the Tyvjobekken Creek and the Reindeer Creek (NW Wedel-Jarlsberg Land, Spitsbergen). We cover hydrometeorological measurements, fluctuations of physicochemical parameters (pH, specific electrolytic conductivity (SEC)), and the presence of selected organic compounds (dissolved organic carbon (DOC), formaldehyde (HCHO), ∑phenols). The obtained levels of DOC (0.061-0.569 mgC $L^{-1}$) and HCHO (<LOD-0.140 mg $L^{-1}$) in water samples of these two high Arctic creeks confirm the role of the melting permafrost as a rich source of terrestrial organic carbon and organic pollutants, as well as the impact of rainfall on surface water chemistry. It was found that fluctuations of physicochemical indices (pH, SEC, DOC) were related to changes in mean daily discharge of Reindeer Creek (0.012-0.034 $m^3$ $s^{-1}$) and Tyvjobekken Creek (0.011-0.015 $m^3$ $s^{-1}$) ($r>0.40$). The Tyvjobekken Creek catchment, in contrast to Reindeer Creek catchment, turned out to be resistant to rapid changes in meteorological conditions ($r <0.10$) and surface runoff. The processes of permafrost thawing, calcium carbonate dissolution, and biogeochemical "breathing" of soils proved to be crucial for the development of water chemistry. In conclusion,

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the surface water chemistry of the Reindeer Creek was found to result from the mutual influence of hydrometeorological indices and the biogeochemical environment of the catchment.

**KEYWORDS:** dissolved organic carbon, formaldehyde, surface water, permafrost, Arctic.

## 1. Introduction

Glaciers cover approximately 60% of the total land area of Svalbard, with the other 40% of the area in the periglacial environment with permafrost (Humlum et al., 2003). The permafrost thickness in Svalbard is estimated to be around 100 m in major valley floors and up to 400-500 m in high mountains (Liestøl, 1976). The published research on the Arctic region confirms that the occurrence, thickness, and thermal state of permafrost depend on many local factors, e.g.: 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). An increase in air temperature over the second half of the 20$^{th}$ century resulted in the widespread permafrost degradation (e.g., Christiansen et al., 2010; Romanovsky et al., 2010; Serreze et al, 2000; Schaefer et al., 2012). Permafrost thawing and an increase in its active layer thickness have also been observed in Svalbard (Isaksen et al., 2003; Christiansen and Humlum, 2008), including Calyspostranda in Bellsund (Marsz et al., 2011). Therefore, the consequence of glacier retreat (Serreze et al., 2000; Flink et al., 2015; Rachlewicz et al., 2016) is the appearance of new ice-free land with favourable conditions for the prevalence of permafrost (e.g. Oliva et al., 2016). The new ice-free environment in the polar zones is characterised by highly dynamic geomorphological and ecological processes (Rachlewicz, 2010; Cooper et al., 2011; Oliva et al., 2016).

The studies related to the permafrost regions show that hydrological processes are controlled by the thickness of the active layer, total thickness of the underlying permafrost, and the distribution of frozen ground and taliks (e.g. Zhang et al., 2002; White et al., 2007; Cheng and Jin, 2012). The hydrogeological conditions of rivers are determined by the freeze-thaw processes in the active layer (Walvoord and Striegl, 2007; White et al., 2007; Ye et al., 2009; Lyon and Destouni, 2010) therefore permafrost thawing may increase river discharges through enhancing infiltration and supporting deeper groundwater flow paths. The occurrence and thawing of permafrost can also influence catchment geochemistry, changing the seasonal fluxes of nutrients, including carbon and nitrogen (e.g., Carey, 2003; Petrone et al., 2006; Larouche et al., 2015). The role of permafrost in the hydrological and hydrochemical processes has yet to be thoroughly understood, particularly in the context of the coexisting glaciers and permafrost thaw. Few works focus on freshwater chemistry in the periglacial zone of the Arctic (e.g. Polkowska et al., 2011; Ruman et al.,2013; Kozak et al., 2015; Rachlewicz et al., 2016) and these show the dependence of the water chemistry on annual changes in precipitation, snow cover and temperature, long distance air transport of contaminants, local geological conditions, and biota.

There is a need for more studies on the influence of seasonal permafrost degradation on the surface water chemistry, and for hydrochemical characteristics of non-glaciated catchments in Svalbard. According to the literature reviewed, the work in this area is scarce and it was mainly carried out within permafrost areas in Siberia (Frey et al., 2007; Bagard et al., 2011) and North America (O'Donnell and Jones, 2006, Douglas et al., 2013). In order to better understand the sources and transport of organic compounds in the Arctic surface waters, we compare the hydrochemistry of two high Arctic creeks with snow-permafrost regime, located on a marine terrace of Spitsbergen. The comparison of hydrochemistry of both creeks permits: 1) to verify how permafrost degradation influences the chemistry of Arctic surface waters, 2) to identify the factors determining differences in hydrological and chemical parameters of the creeks. The first step was to investigate changes in fluctuations of physicochemical parameters (pH, specific electrolytic conductivity (SEC)), dissolved organic carbon (DOC), and formaldehyde (HCHO) in the surface waters of the two non-glaciated catchments. The second step was to define the effect of hydrometeorological conditions on the chemical features of surface waters and to investigate which other factors (e.g. morphology, lithology) could shape water chemistry in both of the studied creeks. Finally, we provide unique information about the loads and transport of organic compounds in the Tyvjobekken Creek and Reindeer Creek. The results presented in this study could also serve as a reference for future changes and the potential effects of advancing permafrost degradation on water chemistry.

## 2. Study area

This surface water study covers two periglacial catchments with snow-precipitation-permafrost alimentation regime, drained by perennial (periodical in their upper courses) creeks (Tyvjobekken and Reindeer Creek), located in the NW part of the Wedel-Jarlsberg Land, in the

Bellsund region of Spitsbergen. The creek valleys have a diverse morphology, from small shallow valleys to gorges. They dissect an area of elevated marine terraces, called Calypsostranda (Fig. 1), which developed as inclined abrasion platforms during glacioisostatic movements in the Younger Pleistocene and Holocene. In Calypsostranda, the terraces develop a system of steps, including the occurrence of berms along the former shorelines, and palaeo-eskers and dead cliffs related to marine abrasion (Landvik et al., 1998).

Both catchments are located on the tectonic units of Calypsostranda Graben, filled with clastic Tertiary sediments. To the layer above 120 m depth is formed by metadiamictice, sandstone type rocks, silicite with Tertiary fossils and loose sandstones, as well as hard coal banks, pebbles and plant remains. Below (to 250 m depth) grey and yellow sandstones can be found, with hard coal remains and sandstone pebbles (Harasimiuk and Gajek, 2013).

In the Tyvjobekken catchment, glacial and fluvioglacial deposits predominate on the surface, without soil cover. Locally, weakly developed loose and alluvial soils occur, while only rarely brown soils can be found there. The soils in the Reindeer Creek catchment are mainly brown soils (rich in organic matter and humus), with a light granulometric composition allowing for the development of a good drainage system. Also, gley soils occur in the upper part of the catchment, while soils are lacking only in the trough of the creek (Fig.6.2. in Klimowicz et al., 2013).

### *2.1. Hydrological characteristics of the studied catchments*

The Tyvjobekken Creek drains the eastern forefield of the Renard glacier, and the slopes of the Bohlinryggen massif, developing a catchment with an area of 1.3 km$^2$. Its springs are located in the forefield of the massif. The Tyvjobekken Creek valley extends from WSW to ENE along a section of approximately 1.2 km, with a mean slope inclination of approximately 4.1% (Kociuba 2015a). Its upper part is fed by a bifurcated stream and has a character of an extensive, weakly developed catchment occupying an area between the moraine zone of the north-eastern Renard glacier forefield and the eastern Bohlinryggen massif. The middle section has permanent drainage, and is fed by the largest (and nameless) tributary, with a length of 350 m and catchment of 0.1 km$^2$. In the middle course, the creek develops a gorge with a depth of up to 25 m (Bartoszewski 1998; Bartoszewski et al., 2013) (Fig.1.C.). That section is characterised by a narrow erosion valley developed by the braided system. In this part, the creek is fed by small periodically functioning tributaries with a snow-permafrost regime. The river bed covers the entire valley floor, and is developed by a single-channel river. The small amounts of water and bedload transported by the Tyvjobekken Creek result in periodical lack of surface inflow of the river to the fjord, and contribute to the development of a microlagoon separated from the waters of the fjord with a coastal berm (Harasimiuk and Król 1992, Kociuba and Janicki 2013, Kociuba 2015a, 2017a).

The Reindeer Creek is the largest tributary of the Scott River, one of the largest proglacial rivers in the study area, flowing directly into the Bellsund Fjord (Kociuba 2017b). This creek also drains an area of approximately 1.3 km$^2$, beginning in the eastern part of the massif. The Reindeer Creek valley extends from SW to NE along a section of approximately 1.3 km, reaching a mean slope inclination of approximately 4.2% (Kociuba 2017c). In the northern part of the catchment, the valley is fed by the waters of a perennial stream with origins at the foot of the eastern slope of the moraines of the Scott Glacier. This tributary feeds the main creek approximately 500 m above the mouth of the Reindeer Creek. The upper section of the Reindeer Creek valley includes two spring niches: its own and of its largest tributary. The valleys are weakly developed here, and the outflow is of discontinuous (declining) character. The middle valley section (approximately 50% of the area of the catchment) is morphologically weakly developed catchment (Fig.1.D.), yet with evident incisions of the main stream and tributary. The drainage is permanent in this section, and the main creek is fed by the largest (nameless) tributary of approximately 450 m length. In the lower course of the Reindeer Creek, a gorge occurs with a depth of up to 25 m (Kociuba, 2015b), where the river bed covers the entire valley floor. Below the gorge, the Reindeer Creek flows into the Scott River.

## 3. Methodology

### *3.1. Sampling and hydrometeorological measurements*

Eighty two surface water samples were collected in total, each assisted with hydrological and meteorological measurements, during 41 days from 13 July to 22 August 2012. Sampling was conducted in the gorge sections of the both creeks located on the Calypsostranda (Fig.1.B). In the field, the personnel taking samples paid extreme attention to avoid contamination, and wore polyethylene gloves. Sampling containers were rinsed with the sample three times, and then filled without air bubble to prevent the loss of analytes to headspace. The study involved the analysis of blank samples to exclude the impact of the containers.

Discharge was measured in gauging sections located in the lower courses of both rivers (Fig.1.B). In the Reindeer Creek, the measurement profile was located 100 m above its mouth to the Scott River, and in Tyvjobekken Creek it was 300 m above the creek's mouth to the Recherchefjord. Water stages in both profiles were recorded 144 times per day with a CTD-Diver meter (Schlumberger Water Services), with a measurement accuracy of ±0.5 cm. The measurement of flow velocity was performed with a current meter HEGA II and an ultrasonic device OTT ADC, with a range of flow velocity measurements of 0.02-3.00 m $s^{-1}$ and 0.2-2.4 m $s^{-1}$ (and an accuracy of ±0.25 cm $s^{-1}$), respectively.

For the measurements of wind directions, air temperature (T), and atmospheric precipitation (P), a portable weather station (Campbell Scientific, CR10X Datalogger) and a Hellmann rain gauge were used. Both the precipitation sampler (with 200 $cm^2$ of inlet ring surface) and the meteorological station were placed approximately 200 m from the seashore at an altitude of 23 m a.s.l. The automatic meteorological station registered data in 10 minute intervals.

### *3.2. Analytical methods*

After collection, the water samples were transported to the laboratory and stored at a temperature of 4°C prior to analysis. The analyses of physical and chemical parameters such as pH and SEC were performed with a microcomputer pH/conductometer CPC-411 by Elmetron, fitted with an EPS-1 electrode and an EC 60 conductivity sensor.

The quantitative analyses of organic compounds were conducted immediately after filtering samples through 0.45 µm. Dissolved organic carbon was determined by catalytic combustion (oxidation), with an NDIR detector. Both the ∑phenols and the formaldehyde (HCHO) levels were determined using spectrophotometry techniques (Table 1).

### *3.3. Quality Assurance/ Quality Control (QA/QC)*

The determination of various targets of analyte groups involved the application of demineralised water type Mili-Q (Mili-Q® Ultrapure Water Purification Systems, Millipore® production). Various matrix compositions of environmental samples require validation of the analytical procedures applied in the determination of individual components against certified reference materials. Moreover, to ensure high quality of results, all data obtained in the research were subject to strict quality control procedures.

### *3.4. Factors for result analysis*

#### *3.4.1. Discharge and load calculations*

The amount of water runoff in the measured profiles of both creeks were determined based on the rating curve (equation 1) (Byczkowski 1999):

$$1. \qquad Q = ah^b$$

where $Q$ is the discharge rate [$m^3\ s^{-1}$], $a$ and $b$ are the curve parameters, and $h$ is the water stage [cm].

The calculation of the chemical compound loads (DOC, ∑phenols, HCHO) from both catchments involved the hydrological-hydrochemical method with the application of the formula (2) after Willson and Bonin (2007) and Buttner (2013).

$$2. \qquad L = C_i Q_i$$

where $L$ is the mean daily load calculated based on concentrations and discharges on the day of sample collection (mg $s^{-1}$), $C_i$ is the concentration of organic compounds in a sample collected on a given day (mg $dm^{-3}$) and $Q_i$ is the mean daily discharge ($m^3\ s^{-1}$).

#### *3.4.2. Statistical methods*

The Student’s t-test and Pearson's correlation coefficients were computed with the software package STATISTICA 6.1 (StatSoft Inc., Tulsa, OK, USA). The significance of differences in the mean of the tested variables (Q, SEC, pH, DOC) between compared creeks was determined by the Student's t-test for 2 independent trials. The calculation of the Pearson's correlation coefficients (r) allows for the detection of pairwise relationships between the meteorological conditions (T, P), mean daily water discharge (Q) and pH, SEC, DOC and HCHO determined in the investigated water samples. Statistical significance of Student's t-test and correlation coefficients was defined at a $p < 0.05$.

# 4. Results

## *4.1. Meteorological conditions*

Meteorological conditions in summer season 2012 in comparison to previous multiannual observations (Mędrek et al., 2014, Franczak et al., 2016), was characterised by lower mean air temperature (4.6°C), lower total precipitation (26.4 mm), and lower mean wind velocity (3.6 m/s).

The prevailing wind directions presented in Fig. 2.A. are also described in detail in Lehmann et al. (2016), and only a brief comparison with the wind conditions in August 2012 (Fig.2.B) will be provided here. Fig. 2.B. shows considerable intensification of winds from WNW and NW (17% and 19%, respectively) in comparison to wind conditions in July presented in the Fig.2.A. In August, we observed the fading of wind coming from the direction of ENE (16% to 2%) and E (10% to 2%), whereas these were among the prevailing wind directions in July. Instead, winds coming from SSE, S, SSW, SW, and W started to prevail (7%, 9%, 11%, 7%, and 7%, respectively). The contributions of the winds coming from the remaining directions (N, NNE, NE, ENE, E, ESE, SE, WSW, NNW) in August ranged between 2% and 4%. During the field sampling in July and August (Fig.2.C), both of the studied catchments were under the influence of the same air masses predominantly coming from WNW and NW (14% and 17%, respectively). Other wind directions, with higher contributions in Fig.2.C., include ENE, E, S, and SSW (8%, 6%, 7%, and 8%, respectively). The Tyvjobekken Creek and Reindeer Creek were hardly ever under the influence of winds coming from N, NNE, NE, ESE, SE, SSE, SW, WSW, W, or NNW (2-5%).

## *4.2. Hydrochemistry characteristics*

Table 2 presents the values of measured hydrological and physicochemical parameters as well as concentration levels of chemical compounds determined in the water samples collected in both catchments.

The frequency distribution of hydrological and physicochemical parameters and organic compound concentrations is shown in Fig. 3. In all samples, the concentrations of phenols were <LOD, hence they are not included in the figure.

The discharges measured in the two creeks differ from each other significantly. Throughout the measurement period in July and August, discharge values in the Tyvjobekken Creek did not exceed 0.016 m $s^{-1}$, while in the Reindeer Creek 63% of measurements were above that value (Fig. 3). Moreover, the fluctuation of water discharges in the Reindeer Creek was almost 6 times greater than in Tyvjobekken. Marked differences also occurred between creeks in the determined values of water pH. In the Tyvjobekken Creek, pH of water samples in 90% varied between 8.00 and 8.20, while for water samples from the Reindeer Creek, such values were recorded only in 12% of the measured samples. In water samples collected from the Reindeer Creek catchment, values between 7.60-7.80 pH and 7.80-8.00 pH were predominant (27% and 54% of measured samples, respectively) (Fig.3).

Specific electrolytic conductivity values <260 µS $cm^{-1}$ in the water collected from Tyvjobekken and Reindeer Creeks were detected rarely (20% and 27%, respectively) (Fig. 3). Values of SEC varying from 260 µS $cm^{-1}$ to 290 µS $cm^{-1}$ as well as 290 µS $cm^{-1}$ to 320 µS $cm^{-1}$ are characteristic of the Tyvjobekken Creek (44% and 29% of results, respectively). In water samples collected from the Reindeer Creek, SEC ranging from 260 to 290 µS $cm^{-1}$ were much less frequent (only 7% of results). Characteristic values of SEC for this creek ranged from 290 to 320 µS $cm^{-1}$, or exceeded 320 µS $cm^{-1}$ (41% and 32%, respectively).

The values of DOC presented in Table 2, differ significantly between the two streams. Both the lowest and the highest values of DOC in the Tyvjobekken Creek are almost twice lower than those determined for the Reindeer Creek. Almost half (44%) of the measured values of DOC in the Tyvjobekken Creek water samples showed concentration <150 mg C $L^{-1}$, while in the Reindeer Creek, the concentration of DOC within that range was determined only in 5% of surface water samples (Fig. 3). In surface water samples from the Tyvjobekken Creek, concentrations of DOC within the range of 0.150-0.200 mg C $L^{-1}$ (39% of measurements) prevailed, while ranges of higher concentration such as 0.200-0.250 mg C $L^{-1}$, 0.250-0.300 mg C $L^{-1}$,and >0.300 mg C $L^{-1}$ occurred less frequently (in 10%, 5%, and 2% of the measured samples, respectively). For surface water samples from the Reindeer Creek, the dominant values of DOC concentration were recorded in the range of 0.190-0.230 mg C $L^{-1}$ (54% of measured samples), while the concentration ranges of 0.150-0.190 mg C $L^{-1}$ and 0.230-0.270 were determined less frequently (17% and 12%, respectively). Values of DOC exceeding the concentration of 0.270 mg C $L^{-1}$ were recorded in 14% of the measured samples. As shown in Table 2 during 41 days of the measurement period, DOC load considerably differed in the Tyvjobekken Creek and Reindeer Creek. Mean daily loads of DOC in the Reindeer Creek (4.97 mg C $s^{-1}$) are more than twice as high as in the Tyvjobekken Creek (2.03 mg C $s^{-1}$).

Formaldehyde, a pollutant with potential carcinogenic and mutagenic properties, was transported by the waters of both creeks. Even though DOC concentrations determined in the Reindeer Creek were often higher than those recorded in the Tyvjobekken Creek, the occurrence of HCHO in this creek was rather rare (Fig. 3). Although HCHO occurred less often in the Reindeer Creek, the value of its maximum concentration was twice as high as that determined in the Tyvjobekken Creek (Table 2). In 90% of surface water samples collected from the Tyvjobekken Creek, HCHO showed values >LOD, while in the Reindeer Creek, HCHO was determined in only 24% of water samples. In the Tyvjobekken Creek, HCHO was recorded predominantly in the concentration range of 0.02-0.06 mg $L^{-1}$ (78%), while in the Reindeer Creek this range corresponded to less than 15% of all samples. However, values above 0.06 mg $L^{-1}$ in both of the creeks occurred similarly in around 10% of the examined samples. In the surface waters of the Tyvjobekken Creek, HCHO occurred almost four times more often than in the Reindeer Creek. The mean loads of HCHO in the studied creeks were 0.539 mg $s^{-1}$ (Tyvjobekken Creek, 37 days) and 1.39 mg $s^{-1}$ (Reindeer Creek, 10 days).

### *4.3. Fluctuations of water discharge*

Based on the conducted hydrometeorological measurements, the fluctuation of water discharge in both creeks on the background of changing meteorological conditions is presented. In the Fig.4., two variables are shown with a possible influence on the fluctuation of water discharge: mean air temperature and rainfall.

In the Tyvjobekken Creek, a systematic decrease in discharges occurred since the beginning of the measurement period, from the maximum value of 0.015 $m^3$ $s^{-1}$ on 13th July to a minimum of 0.011 $m^3$ $s^{-1}$ on 7th August. The mean discharge in the entire study period of 2012 amounted to 0.013 $m^3$ $s^{-1}$, which corresponds to a total runoff of 46 000 $m^3$ and a runoff layer of 35 mm. The discharge of the Reindeer Creek, however, was characterised by a marked variability. At the beginning of the ablation season, two maximum values of discharge were recorded amounting to 0.034 $m^3 \cdot s^{-1}$. The mean discharge in the Reindeer Creek amounted to 0.021 $m^3 \cdot s^{-1}$. The value corresponds to the total runoff of 73 000 $m^3$ and a runoff layer of 56 mm (Fig.4).

Daily air temperature fluctuations and precipitation events presented in Fig. 4. show no influence on discharge in the Tyvjobekken Creek, whereas rapid increases in discharge are observed in the Reindeer Creek in response to precipitation events.

*4.4. Temporal variations in hydrochemistry*

Fig. 5. A.-F. presents the temporal and spatial distribution of the pH, SEC, DOC, and HCHO concentration on the background of the water discharge in the both creeks.

As presented in Fig. 5.A.B., on the first days of the research (13th-21st July), in both of the studied catchments, marked increases in pH and SEC were observed. In the Tyvjobekken Creek, pH increased over only three days from 7.40 to 7.94. In the Reindeer Creek during the same time, pH increased rapidly from 7.26 to almost 7.79. During the rest of the summer season, pH values in the Tyvjobekken Creek were stable, and oscillated between 8.00 and 8.20 pH independently of the occurrence of precipitation events or temperature changes. pH values in the Reindeer Creek during the rest of the summer season were much more variable, from 7.67 to 8.09. On 22nd July, the only case of water pH decrease in response to the occurrence of a heavy rainfall event (11.4 mm) was recorded (Fig. 5.B).

Fig. 5.C.D. shows a rapid increase in SEC values on 21st July in both of the creeks. In the Tyvjobekken Creek, an increase in SEC of almost 50 $\mu S\ cm^{-1}$ occurred from 196 $\mu S\ cm^{-1}$ to 245 $\mu S\ cm^{-1}$, while in the Reindeer Creek, an increase in SEC of approximately 100 $\mu S\ cm^{-1}$ was observed, from 205 $\mu S\ cm^{-1}$ to 297 $\mu S\ cm^{-1}$. On 21st July, an event of heavy rain began which lasted until 23rd July. During that time, 15.5 mm of precipitation fell. SEC values in both of the creeks increased gradually until the end of the measurement period. A gradual increase in SEC accompanied by a slow decrease in water discharge (Fig. 5.C.D) indicates high importance of water discharge for SEC values in surface waters. However, on 21st July, no rapid changes in the water discharge of the Tyvjobekken Creek or the Reindeer Creek were observed that could explain the sudden increase in SEC values in surface waters on that day.

The transport of organic compounds in each of the creeks was different (Fig. 5.E.F.). Dissolved organic carbon levels in the Reindeer Creek were almost twice as high as in the Tyvjobekken, where the presence of organic carbon in surface waters included HCHO occurring almost every day. A decrease in the water discharge was accompanied by an increase in DOC levels (Fig. 5.E). However, in the Reindeer Creek, throughout the measurement period, DOC levels oscillated around 0.200 mgC $L^{-1}$. From $21^{st}$ to $23^{rd}$ July, as well as on days when precipitation events occurred and water discharge increased, also an increase in DOC concentration was observed (Fig. 5.F). In the beginning of August (from $6^{th}$ to $10^{th}$ August), the presence of HCHO was recorded, simultaneous with a precipitation event and the lowest levels of water discharge across the whole measurement period. In the following days, when water discharge in the Reindeer Creek increased, an increase in DOC concentration was also observed. HCHO occurred in the waters of the Reindeer Creek more often when water discharge was below 0.100 m $s^{-1}$.

## 4.5. *Correlation analysis*

The degree of correlation between the obtained data and hydrological and meteorological parameters was interpreted as follows: +/- ($r>0.50$) - no justification for rejecting the significant correlation hypothesis; +/- ($r=0.30$-$0.50$) - suspected correlation; +/- ($r<0.30$) - no correlation (Stanisz 1998). The correlation matrix of the analysed indices (Table 3.A.B) showed significant negative correlations between Q (discharge) and pH and SEC in both creeks, as well as the DOC concentration (only in the Tyvjobekken Creek), while a strong positive correlation was found between Q and DOC in the Reindeer Creek. Meteorological variables showed a moderate positive correlation between temperature (T) and precipitation (P) ($r=0.38$); T also showed a weak positive correlation with Q in the Reindeer Creek ($r=0.24$) and HCHO concentration in the Tyvjobekken Creek ($r=0.21$). A moderate positive correlation was recorded between pH and SEC in both the Tyvjobekken and the Reindeer Creek ($r=0.51$ and $r=0.40$, respectively). A significant negative correlation occurred between SEC and DOC in the Reindeer Creek ($r=-0.34$) while a weak positive correlation occurred between these variables in the Tyvjobekken Creek ($r=0.20$). In conclusion, no significant correlations were observed between meteorological parameters: P (precipitation), T (temperature), and Q (discharge) as well as with other analysed physical-chemical indices.

## 5. Discussion

### *5.1. Morphological factors of the catchment affecting water hydrochemistry*

The thickness of the permafrost active layer on Calypsostranda in the catchment areas of the Tyvjobekken Creek and Reindeer Creek in 2012 was at the same level (>202cm) (Repelewska-Pękalowa et al., 2013). According to AMAP report (2012), permafrost affecting water storage and stream runoff responds very slowly to rapidly changing climate conditions. That explains very small oscillation of water discharge in the Tyvjobekken catchment, characterised by almost no response to changes in meteorological conditions. Conversely, the considerable variations of water discharge in the Reindeer Creek were caused mainly by intensive surface runoff of rain water.

Both studied creeks are fed in summer by thawing permafrost waters, precipitation, and snow cover (mainly during spring) (Bartoszewski et al., 2013). Water discharges presented in this paper concerning both of the creeks differ significantly despite their similar alimentation regime, catchment area and mean inclination. The discharge in the Tyvjobekken Creek is regular and half of the level found in the Reindeer Creek. It responds neither to mean air temperature changes (r= 0.07) nor occurrence of precipitation (r=0.07). Meanwhile, water discharge in the Reindeer Creek responds to changes in temperature, and temporally increases with each precipitation event. Although the effect of rainfall on the hydrological conditions of the Reindeer Creek is not confirmed by the correlation matrix results presented in Table 3 (r= 0.07), it cannot be excluded, considering even 50% errors related to rainfall collection in Svalbard, caused by: high wind speed, open tundra and non-representative locations for precipitation stations (Killingtveit et al.,2004). Throughout the measurement period, both catchments were under the influence of the same meteorological conditions. The dissimilarity of the hydrology between the studied creeks results from morphological differences of their catchments and differentiation of their soil formation. Shape of the Tyvjobekken catchment is irregular with a poorly developed network of watercourses in its upper part. Moreover, the catchment is rich in numerous small tundra lakes which impede surface runoff. Additionally, the scarcity of soil cover, poor vegetation in this part of Calypsostranda, and the dry surfaces of marine terraces favour the evaporation of water which reaches this catchment through rainfall (Klimowicz et al., 2013; Repelewska-Pękalowa et al., 2013). Changes in water discharge in the Tyvjobekken Creek during the 1987 hydrological year were described by Bartoszewski et al. (2013). In comparison to their description, in 2012 the studied water discharges and chemical composition in the Tyvjobekken Creek were particularly related to the thawing of permafrost. Shape of the Reindeer Creek valley is close to a trough. It is wider in the upper part of the catchment, and due to the domination of brown soils has a well-developed network of watercourses which provide water also from the Scott Glacier terminal moraine where gley soils dominate. These factors favour easier surface runoff from the entire area of the catchment, and result in higher water discharge in the creek. According to Repelewska-Pękalowa et al. (2013), in the area of the Reindeer Creek catchment, zones of active solifluction and periodically wet terraces predominate, favouring accumulation of water and its easier release in response to rainfall events. This suggests that the hydrochemistry of the Reindeer Creek waters was determined by rainfall, thawing of permafrost, and melting of the snow cover from the area of the glacier terminal moraine.

*5.2. Hydrochemistry and loads of organic compounds*

The pH values in the Tyvjobekken Creek and Reindeer Creek correlate negatively with water discharge. A similar effect is even more striking for SEC values. This correlation between the hydrology of non-glaciated streams and the chemistry of their waters was also pointed out by Chmiel et al. (2013). Repelewska-Pękalowa and Magierski (1989) found that changes in SEC values of non-glaciated streams are related to an additional load of waters coming from thawing of permafrost and the cryochemical effect. This explains the fluctuations of SEC in the Tyvjobekken Creek and the results of the matrix correlation analysis showing a negative correlation with water discharge. More varied fluctuations of pH and SEC values in the Reindeer Creek during the summer season of 2012 are most likely related to an increase in the contribution of water coming from other sources than permafrost thawing (e.g. rainfall, snow cover). According to Kozak et al. (2015) and Lehmann et al. (2016), the pH of rain in Svalbard ranges from 4.43 to 7.93 pH, and could possibly affect the pH of surface water. One of the main factors determining the chemistry of surface waters in Svalbard is rock-water interaction such as dissolution of calcium carbonate (Stutter and Billet, 2003; Dragon and Mariciniak, 2010; Chmiel et al., 2013) which is responsible for the alkaline character of the surface waters. Less alkaline character of the Reindeer Creek in comparison to the Tyvjobekken Creek may be explained by the vulnerability of this catchment to rainfall and the domination of brown soils which are a source of humic acids. A noticeable moderate positive correlation between pH and SEC, found in both the Tyvjobekken Creek and Reindeer Creek is related to the biogeochemical factor. The aforementioned process of calcium carbonate dissolution increases water pH and SEC values, and may be understood as the chemical factor here. According to Chmiel et al. (2013), the alkaline character and the higher concentration of ions in water in the non-glaciated areas of Svalbard, such as the Tyvjobekken Creek and Reindeer Creek, is also determined by a biotic factor ("breathing" of biogeochemicals in soils).

The occurrence of organic pollutants in the Arctic may be a consequence of their long range atmospheric transport (LRTAP) from industrialised and urbanised areas of Eurasia (Hallanger et al., 2011; Kozak et al., 2013). Xu et al. (2016) point to South and East Asia as the main sources of black carbon (BC) and organic carbon (OC) in the world. Studies of Ruman et al. (2014) and Kozak et al. (2015) prove that wet precipitation is a source of organic compounds (TOC, HCHO, ∑phenols) in Svalbard in each season of the year. However, according to AMAP (2012), terrestrial permafrost is also a rich source of carbon. Chmiel et al. (2013) suggest that high values of organic compounds in peat bogs surrounding the Reindeer Creek correspond with high levels of nitrogen and phosphorus indicating their local biological source (bird colonies and reindeer herds in the vicinity of the creek).

According to Weishaar et al. (2003), a vast amount of organic matter in water samples has the form of DOC. Arctic surface water can transport large amounts of organic compounds from the terrestrial environment to neighbouring seas and fjords (Büttner and Tittel, 2013; Dittmar and Kattner, 2003). The DOC determined in the surface waters of Tyvjobekken corresponds negatively with Q, while the DOC determined in the Reindeer Creek has a strong positive correlation with Q. A negative influence of Q on DOC in Tyvjobekken is particularly noticeable in July ($13^{th}$-$25^{th}$ July). Higher water discharge during that time could be related to melting of the snow cover or to a more intense thawing of permafrost. Both of these process result in an additional load of fresh water, and a dissolution of organic compounds flushed out from the soil. Based on the correlation matrix analysis, DOC determined in water samples from the Tyvjobekken Creek, as well as pH and SEC, was related to the biogeochemical processes occurring in soils.

Dissolved organic carbon in the Reindeer Creek catchment was positively correlated with Q. The highest concentration of DOC (0.569 mg C $L^{-1}$) was a result of the occurrence of a heavy rain event (21st-23rd July). The Reindeer Creek catchment is rich in wet tundra, and both plant vegetation and the activity of reindeer in that area is important. It has a direct effect on the high levels of DOC in the creek water. Moreover, easy surface runoff also favours flushing of organic pollutants which reach the catchment through wet and dry deposition. Dissolved organic carbon indices correspond negatively with geochemical conditions of the environment. This suggests the importance of the activity of reindeer herds and surface runoff as the main factors determining the level of DOC in the Reindeer Creek.

Taking into consideration that plant vegetation, the activity of reindeer herds, and the intensity of surface runoff are poorer in the Tyvjobekken catchment than in the Reindeer Creek catchment, the DOC level in the Tyvjobekken Creek results particularly from biogeochemical processes in the soil, and less from surface runoff or wet and dry deposition, while the origin of the DOC in the Reindeer Creek is much more complex.

Studies of Ruman et al. (2014) confirm the presence of HCHO and phenols in precipitation water collected during all four seasons of the year in Svalbard (reporting levels of 0.025-0.150 mg $L^{-1}$ and 0.025-0.075 mg $L^{-1}$, respectively). This suggests constant inflow of these contaminants to the Arctic. However, no phenols were determined in the surface water samples from either of the two sampled creeks. The correlation matrix results do not point to the influence of any variables mentioned herein on the occurrence of HCHO in both creeks. A small amount of rain has no effect on the hydrochemistry of the Tyvjobekken Creek, and has little impact on Q and pH in the Reindeer Creek. Therefore, HCHO determined in the waters of the Tyvjobekken Creek is most certainly related to the thawing of permafrost and biogeochemical processes occurring in soils (F2). The occasional occurrence of HCHO in the Reindeer Creek is related to the dilution of HCHO from thawing permafrost by the water coming from surface runoff or rainfall. HCHO determined in the Tyvjobekken Creek may be identified as being of local origin. HCHO occurs in the Reindeer Creek particularly when the discharge is lower (26th July to 17th August), and when rainfall occurs (7th-10th August). Considering the natural sources of HCHO and the increased plant vegetation and reindeer herds activity in the Reindeer Creek catchment, it cannot be excluded that HCHO present in its water origin from natural sources as well as from LRTAP.

The transport of organic compounds from the Tyvjobekken Creek catchment is more difficult than in the case of the Reindeer Creek due to the morphological aspects of the area (see section

5.1). However, next to morphological aspects favouring surface runoff, higher loads of organic compounds in the surface waters of the Reindeer Creek are also largely determined by the soil type, vegetation cover in the area of the catchment, and the related activity of animals. Presented results show that even low levels of chemical compounds determined in the Tyvjobekken and Reindeer Creek could correspond to relatively high levels of loads of such substances due to the discharge of the creeks. The calculated loads of chemical compounds show what amounts of pollutants may actually reach Arctic fjords as a result of permafrost thawing.

## 6. Conclusions

Hydrometeorological research showed that 4.6°C of mean air temperature was enough to have influence on permafrost degradation in Calypsostranda and the provision of melt waters to feed both creeks during all summer season in year 2012. Based on the conducted chemical analyses, we were able to conclude that thawing permafrost is a source of dissolved organic carbon, including formaldehyde, which was clearly visible in water of the Tyvjobekken Creek. A further rise in air temperature in the Arctic may result in an intensification of permafrost degradation which would lead to changes in surface water discharge, and release higher loads of HCHO, which due to its carcinogenic and mutagenic properties may negatively impact the Arctic environment.

The correlation matrix analysis confirms an important role of rock-water interaction in shaping the chemistry of High Arctic surface waters. The presented study proves also that the influence of permafrost degradation and rainfall on surface water hydrochemistry in a non-glaciated Arctic catchment, depends significantly on the morphology of catchments as well as the types of soils covering them. The predominance of brown soil in the Reindeer Creek catchment resulted in a better developed water drainage, a lush plant cover and a more intense activity of reindeers in this area in comparison to Tyvjobekken catchment. These factors combined directly affect the differences in hydrochemistry of the creeks.

In conclusion, next to atmospheric deposition and rock-water interaction, the crucial influence on surface water hydrochemistry of a non-glaciated Arctic catchment in Svalbard, is also exerted by: the rate of permafrost degradation, the geomorphology of the catchment, the dominating types of soils, the presence of vegetation and animal activity.

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