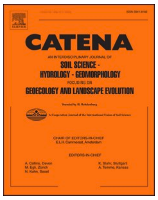
Catena 231 (2023) 107311

Contents lists available at ScienceDirect

Catena

journal homepage: www.elsevier.com/locate/catena



Biogeochemical cycling in a periglacial environment – A multi-element mass-balance budget for a catchment in West Greenland

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ARTICLE INFO

Keywords:
Multi element
Mass-balance budget
Whole catchment
Dry periglacial landscape
Terrestrial
Aquatic

ABSTRACT

There is an increased awareness that the biogeochemical cycling at high latitudes will be affected by a changing climate. However, because biogeochemical studies most often focus on a limited number of elements (i.e., C, P and N) we lack baseline conditions for many elements. In this work, we present a 42-element mass-balance budget for lake dominated catchment in West Greenland. By combining site specific concentration data from various catchment compartments (precipitation, active layer soils, groundwater, permafrost, lake water, lake sediments and biota) with catchment geometries and hydrological fluxes from a distributed hydrological model we have assessed present-day mobilization, transport and accumulation of a whole suite of elements with different biogeochemical behavior. Our study shows that, under the cold and dry conditions that prevails close to the inland ice-sheet: i) eolian processes are important for the transport of elements associated with mineral particles (e.g., Al, Ti, Si), and that these elements tend to accumulate in the lake sediment, ii) that even if weathering rates are slowed down by the dry and cold climate, weathering in terrestrial soils is an important source for many elements (e.g., lanthanides), iii) that the cold and dry conditions results in an accumulation of elements supplied by wet deposition (e.g., halogens) in both terrestrial soils and the lake-water column, and iv) that lead and sulfur from legacy pollution are currently being released from the terrestrial system. All these processes are affected by the climate, and we can therefore expect that the cycling of the majority of the 42 studied elements will change in the future. However, it is not always possible to predict the direction of this change, which shows that more multi-element biogeochemical studies are needed to increase our understanding of the consequences of a changing climate for the Arctic environment.

1. Introduction

Arctic environments are experiencing rising temperatures at an unprecedented rate (Box et al., 2019; IPCC, 2013), and in the last few decades there has been an increasing number of studies discussing how a warmer climate will affect biogeochemical processes at high latitudes (Anderson et al., 2017; Hinzman et al., 2013; Hobbie et al., 2017; Lecher, 2017; Myers-Smith et al., 2020; Pokrovsky et al., 2012; Pold et al., 2021; Schuur et al., 2015; Smol, 2012; Vonk et al., 2015; Zimov et al., 2006). However, the majority of these studies only concern a single element (e.g., carbon) or a small selection of elements (e.g., carbon, nitrogen and phosphorus). Therefore, for many elements, there is a lack of region-specific baseline conditions that future changes can be compared against (Anderson et al., 2017). Considering the complex nature of Arctic environments, such baseline conditions should preferably be determined on a landscape scale, integrating both the terrestrial and aquatic systems (Hanson et al., 2015; Solomon et al., 2015; Umbanhowar et al., 2015).

The Arctic is different from other regions in several ways. The presence of permafrost restricts the movement of water through deeper soil layers and can result in a limited hydrological connectivity between the terrestrial and aquatic systems (Jessen et al., 2014; Lecher, 2017).

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https://doi.org/10.1016/j.catena.2023.107311
Received 18 January 2023; Received in revised form 21 April 2023; Accepted 13 June 2023

Increased permafrost thaw can therefore lead to a mobilization of elements currently stored in terrestrial soils, resulting in an increased loading of, e.g., mercury to adjacent lakes (Rydberg et al., 2010). A warmer climate might also affect the rate of organic matter degradation, soil formation, and chemical weathering, due to faster process rates (Goudie and Viles, 2012; Hall et al., 2012; Laudon et al., 2012; White and Blum, 1995). At the same time as the weathering rate increases, there will – for areas close to an inland icesheet – be a reduced eolian input of fresh, unweathered, material from glacial outwash plains (Bullard and Austin, 2011; Rydberg et al., 2016; Willemse et al., 2003). This implies that even with faster weathering rates, it is not certain that a warmer climate will lead to an increased release of weathering products (Fritz et al., 2004; Scribner et al., 2015).

One tool for assessing present-day biogeochemical cycling of elements is to compare the input and output for a specific system. Over the years, such mass-balance approaches have been successfully used to study various processes, including weathering, cation exchange and soil formation (Cornwell, 1992; Hood et al., 2019; Huang et al., 2013; Land and Ohlander, 2000; Velbel and Price, 2007), the effects of landscape development (Chamberlin et al., 2019), carbon and nitrogen cycling (Bormann et al., 1977; Hanson et al., 2015; Lindborg et al., 2020; Lundin et al., 2016), and transport and retention of anthropogenic pollutants (Graydon et al., 2012; Lankey et al., 1998; Nelson et al., 2007). Studies in the fields of petrology and geology tend to use mass-balance budgets that include more than ten elements (Li et al., 2020; Velbel and Price, 2007), while most studies within the field of biogeochemistry rely on a single or small selection of elements (Johnson et al., 2000; Lundin et al., 2016; Mitchell et al., 1996; Navrátil et al., 2007; Oulehle et al., 2019). Including a larger number elements can have several advantages for biogeochemical studies. First, utilizing multiple elements can reduce the sensitivity to spatial and temporal variability, as well as analytical uncertainties. Second, because of differences in their biogeochemical cycling, using more elements can simultaneously assess a broader spectrum of processes, sources and sinks.

Halogens such as chlorine (Cl), bromine (Br), and iodine (I) primarily enter the landscape through atmospheric deposition and have a predominate marine origin (Duce et al., 1965; Willis et al., 2018). Because of their high water-solubility they tend to be relatively mobile, and Cl is often used as a conservative element to trace solute transport though the landscape (Lockwood et al., 1995). Atmospheric deposition is also a source for alkali and alkaline earth metals (e.g., Na, K, Ca) and various metals and metalloids (e.g., Cu, Zn, Bi, Mo, Sb, S, Pb; Heidam et al., 2004; Pearson et al., 2019; Steinnes et al., 2011). Unlike the halogens, many of these elements readily sorb to soil particles (especially organic matter), or are plant nutrients (e.g., K, P) that are actively taken up by plants (Johnson et al., 2000; Peltola et al., 2008; Raulund-Rasmussen et al., 1998). This means that they can be used to assess the accumulation in and release from terrestrial soils, plants and decaying organic matter (Biester et al., 2012; Lidman et al., 2014; Tiberg et al., 2018).

In addition to the atmospheric inputs, weathering of silicate and carbonate minerals is an important internal source for several elements (Hu and Gao, 2008; Raulund-Rasmussen et al., 1998). Calcium is released during both carbonate and silicate weathering, but a high release of Ca (together with Sr) from the terrestrial system is an indication of carbonate weathering (Bailey et al., 1996). Silicate weathering is generally much slower, but it still results in the mobilization of a whole suite of elements (Minarik et al., 1998; Tyler, 2004; Velbel and Price, 2007). Some of these weathering products are easily dissolved in water and are relatively mobile (e.g., K, Ca, Li), while others either require certain pH or redox conditions to stay in solution (e.g., Al, U, Mo, V, B, Fe) or are dependent on the complexation with dissolved organic matter (DOM) or other colloidal phases to become mobile (e.g., Lanthanides; Bailey et al., 1996; Hazen et al., 2013; Robinson et al., 2018; Saleh et al., 2018; Tyler, 2004). Finally, even if there is some release of elements like, e.g., Si, Ti and Zr, during weathering, they tend to become enriched in the residual mineral material, and can therefore be used as tracers for the transport of particulate material (Hazen et al., 2013; Rydberg et al., 2016; Taboada et al., 2006). Taken together, this variability in sources, mobility, and tendencies to accumulate (or become depleted) in different landscape compartments provides an opportunity to assess a whole range of biogeochemical processes on a landscape scale.

In this study, we set up a mass-balance budget for 42 elements that provide estimates for the major pools and fluxes for the entire Two-Boat Lake catchment in West Greenland (Tröjbom and Grolander, 2010; Velbel and Price, 2007). Our overarching aim was to improve the understanding of biogeochemical cycling in cold and dry periglacial landscapes. In particular, our study was intended to answer three more specific questions: i) How does the combination of low temperatures and young, undeveloped, soils affect the release of weathering products, ii) What are the consequences of the restricted hydrological transport for the retention and transport of easily dissolvable elements (e.g., halogens) supplied primarily by wet deposition, and iii) Are there any signs of release of legacy pollutants (e.g., Pb) at this remotely located site. This knowledge is then used as a basis for a discussion of how the biogeochemical cycling would change under a future warmer climate.

2. Methods

2.1. Site description

The Two-Boat Lake catchment (TBL; Lat 67.126 Long − 50.180; Fig. 1) is situated 25 km northeast of Kangerlussuaq in West Greenland (others have also referred to the lake as SS903, e.g., Sobek et al., 2014). Regional bedrock is dominated by tonalitic and granodioritic gneisses (van Gool et al., 1996), and quaternary deposits are mainly made up of till or glacifluvial deposits that are overlain by eolian material (Petrone et al., 2016). The TBL catchment has continuous permafrost (estimated to reach down to 400 m), which is interrupted by a through talik – unfrozen ground underneath the lake – that connects the lake with the regional groundwater system (Harper et al., 2011). Despite the proximity to the ice sheet (about 500 m) the catchment receives no meltwater from the ice sheet, and it has been deglaciated for at least 4000 years (Levy et al., 2012; Lindborg et al., 2016; Rydberg et al., 2016; van Tatenhove et al., 1996; Young and Briner, 2015). Based on the geology, vegetation, hydrology and lake water chemistry, the TBL-catchment can be regarded as being representative of the periglacial region close to the Greenland icesheet (Henkemans et al., 2018; Johansson, 2016; Lindborg et al., 2016; Ryves et al., 2002; Sobek et al., 2014).

During the period 2010–2019, the Greenland Analogue Surface Project (GRASP) conducted a thorough survey of the hydrology and geohydrology of the TBL catchment. Petrone et al. (2016) report that most of the catchment is covered by eolian deposits that are, on average, one meter thick, that terrestrial vegetation can be divided into dwarf shrub, meadow, dry grass and wetland communities, and that the active layer is between 0.5 and 0.8 m thick depending on vegetation type. By using site specific data (meteorology, soil temperature, soil moisture, potential evapotranspiration, hydraulic conductivity, and soil porosity) together with a detailed digital elevation model and a 3D model of the active layer depth and soil stratigraphy (Petrone et al. 2016), Johansson et al. (2015b) developed a distributed and physically based hydrological model specifically for the TBL-catchment using MIKE SHE. This model enables temporally and spatially resolved quantifications of both surface and groundwater flow into the lake, both for the entire catchment and for different sub-catchments. The model was calibrated against observed variations in lake water level, groundwater level in the active layer, groundwater transport in the underlying talik and soil moisture, as well as, the annual-water balance for the TBL-catchment for the period 2010–2013. In order to ensure that the relatively short calibration period was representative of the long-term climate the meteorological data for 2010–2013 was validated against a regional climate series covering the period 1977–2013. In brief, the model gives that of the

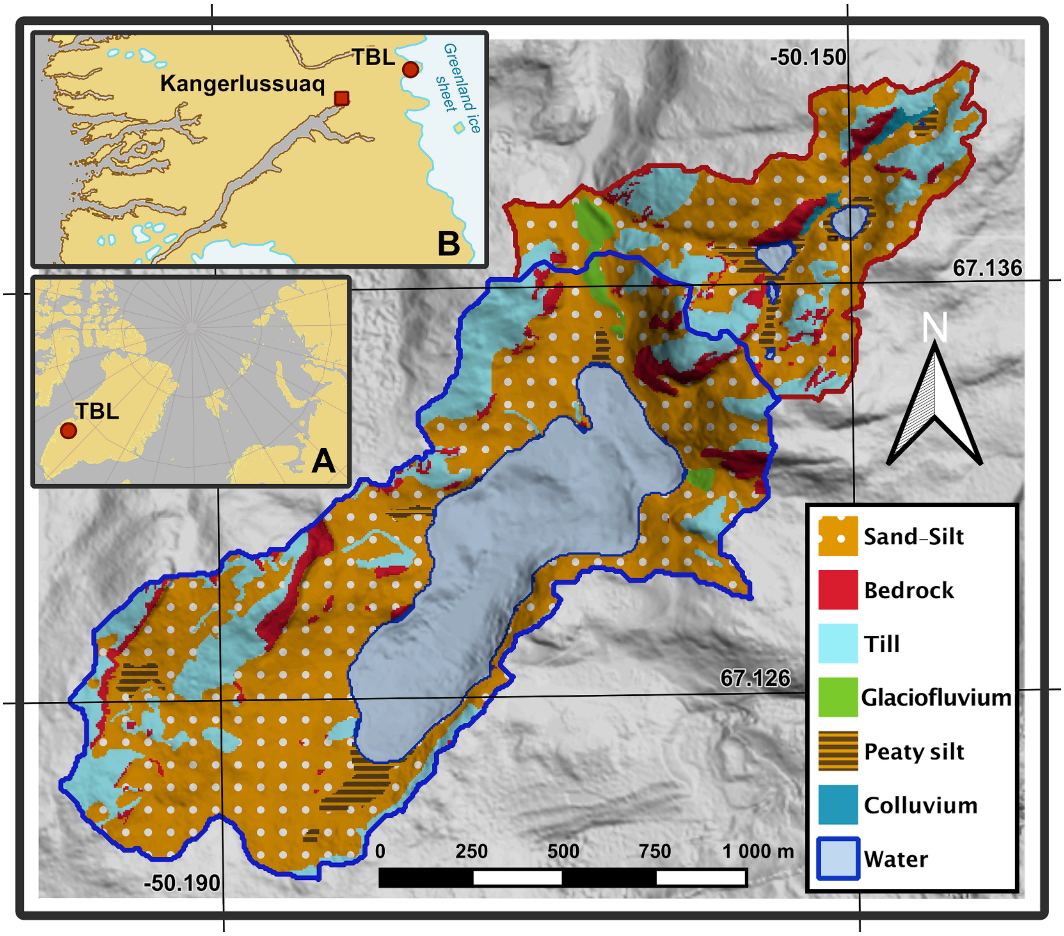


Fig. 1. Maps showing the location of the TBL catchment (inserts A and B), and the distribution of quaternary deposits within the catchment. The total catchment area is 1.7 km^2, and the blue line marks the originally defined catchment area (1.2 km^2; as reported by, e.g., Petrone et al., 2016), while the red outline delimits the additional area that contributes with runoff to the lake during the snowmelt period (0.5 km^2).

annual precipitation during a representative normal year (269 mm; defined as 2013 based on weather data for the period 1977–2013) 40% falls as snow, 52% of the input of water to the lake occurs during the snowmelt period, the potential evapotranspiration is 400 mm, and the runoff from the terrestrial system to the lake is 70 mm (SI-Table 1). Furthermore, there are no permanent streams, but small temporary "streams" appear during very wet periods, and outflow from the lake occurs only sporadic.

Based on the flow of water during the period from late June to September, previous studies reported the terrestrial catchment area as 1.2 km^2 (Johansson et al., 2015b; Lindborg et al., 2020; Petrone et al., 2016; Rydberg et al., 2016). Observations, made in late May to early June (in 2016 and 2017), revealed that an additional area to the northwest of the original catchment area also contributes to the runoff during the snowmelt period (i.e., late May to mid-June; Fig. 1). Redefining the catchment boundary, gives a total terrestrial area of 1.7 km^2. The added northeastern part (0.5 km^2) is, however, disconnected from the main catchment for most part of the year, and only contributes to the runoff until the deeper sections of the active layer becomes unfrozen in mid-June.

2.2. The mass-balance budget

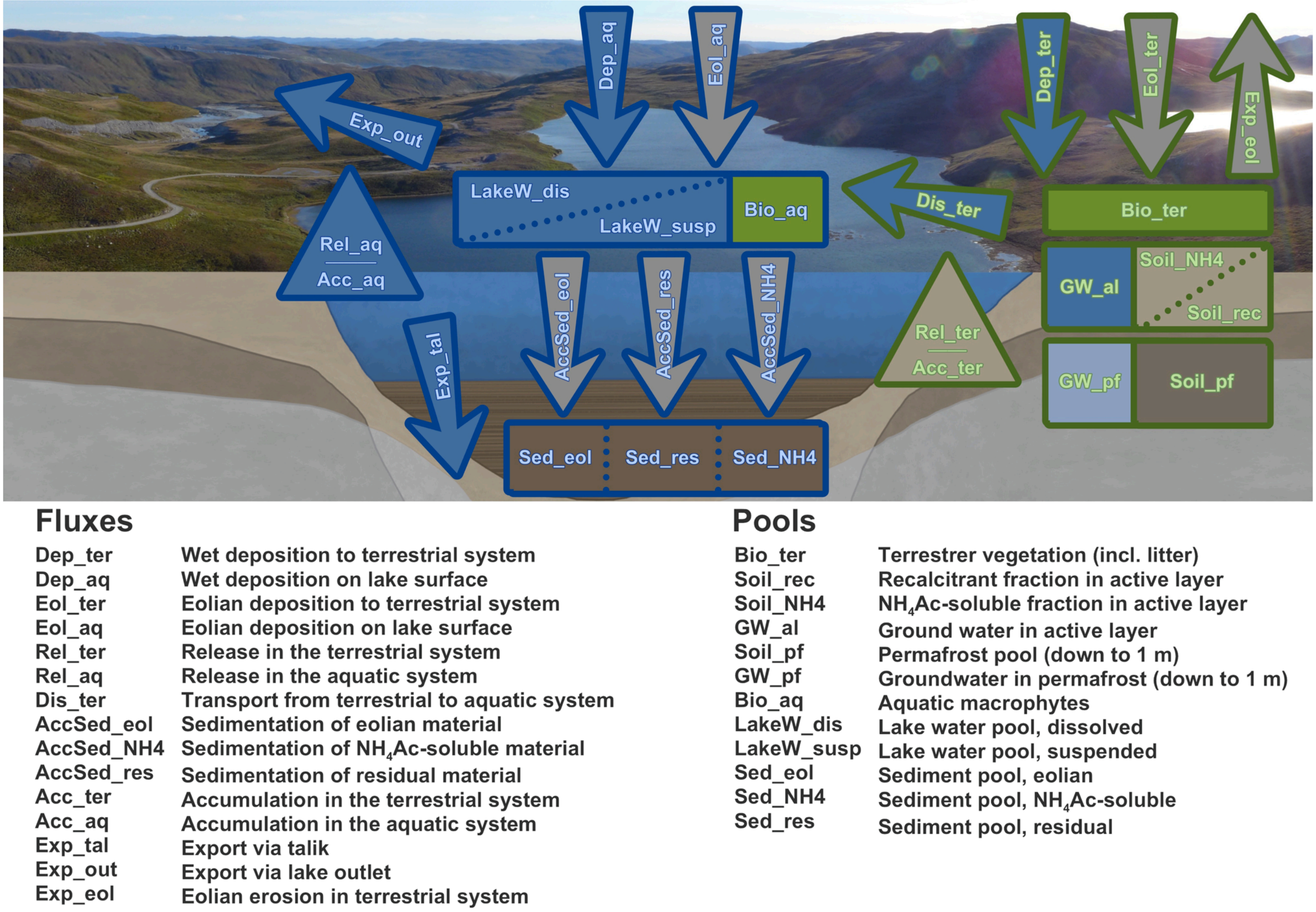
The mass-balance budget can be broken down into a number of pools (i.e., terrestrial biota, active layer, permafrost, aquatic biota, lake water and lake sediment) and the fluxes in and out of them (Fig. 2). Primary input to the system occurs as wet deposition (precipitation) and eolian deposition, whereas export occurs as eolian erosion in the terrestrial system and lake-water outflow (via the lake outlet and through the talik). Internal fluxes include, e.g., hydrological transport with runoff to the aquatic system and burial in lake sediments, which together with the primary input and export result in changes in the storage of elements within the different catchment pools (Fig. 2).

In the mass-balance budget one equation describes fluxes in and out of the terrestrial system (Eq. (1), while a second equation describes fluxes in and out of the aquatic system (Eq. (2)).

$$\mathrm{Dep\_ter} + \mathrm{Eol\_ter} + \mathrm{Rel\_ter} = \mathrm{Acc\_ter} + \mathrm{Dis\_ter} + \mathrm{Exp\_eol} \tag{1}$$

$$\begin{aligned}\mathrm{Dis\_ter} + \mathrm{Dep\_aq} + \mathrm{Eol\_aq} + \mathrm{Rel\_aq} = {} & \mathrm{Exp\_tal} + \mathrm{Exp\_out} + \mathrm{AccSed\_eol} \\ & + \mathrm{AccSed\_NH4} + \mathrm{AccSed\_res} \\ & + \mathrm{Acc\_aq}\end{aligned} \tag{2}$$

Dep_ter and Dep_aq are the inputs as wet deposition on the terrestrial and aquatic systems, respectively; Rel_ter is the release of elements from the terrestrial system; Acc_ter and Acc_aq are the accumulation of elements in the terrestrial system and lake water, respectively; Dis_ter is the transport from the terrestrial to the aquatic system with runoff; Eol_ter and Eol_aq are the inputs of eolian material to the terrestrial and aquatic systems, respectively; Rel_aq is the release of elements in the aquatic system; Exp_eol is the export of material from the terrestrial system through eolian activity; Exp_tal and Exp_out are the export out of the system through the talik and outlet, respectively; AccSed_eol, AccSed_NH4 and AccSed_res are the accumulation in lake sediments



Fluxes

Dep_ter	Wet deposition to terrestrial system
Dep_aq	Wet deposition on lake surface
Eol_ter	Eolian deposition to terrestrial system
Eol_aq	Eolian deposition on lake surface
Rel_ter	Release in the terrestrial system
Rel_aq	Release in the aquatic system
Dis_ter	Transport from terrestrial to aquatic system
AccSed_eol	Sedimentation of eolian material
AccSed_NH4	Sedimentation of NH_4Ac-soluble material
AccSed_res	Sedimentation of residual material
Acc_ter	Accumulation in the terrestrial system
Acc_aq	Accumulation in the aquatic system
Exp_tal	Export via talik
Exp_out	Export via lake outlet
Exp_eol	Eolian erosion in terrestrial system

Pools

Bio_ter	Terrestrer vegetation (incl. litter)
Soil_rec	Recalcitrant fraction in active layer
Soil_NH4	NH_4Ac-soluble fraction in active layer
GW_al	Ground water in active layer
Soil_pf	Permafrost pool (down to 1 m)
GW_pf	Groundwater in permafrost (down to 1 m)
Bio_aq	Aquatic macrophytes
LakeW_dis	Lake water pool, dissolved
LakeW_susp	Lake water pool, suspended
Sed_eol	Sediment pool, eolian
Sed_NH4	Sediment pool, NH_4Ac-soluble
Sed_res	Sediment pool, residual

Fig. 2. Conceptual illustration of the mass-balance budget. Pools are drawn as boxes, fluxes as arrows and the triangles represent the balancing terms used to represent internal sources and sinks. Green outlines denote pools and fluxes in the terrestrial system, while blue outlines relate to the aquatic system. The colour of boxes indicates the type of pool, i.e., biota (green), water (dark blue), ice (light blue), active layer soil (light brown), and the less mobile pools in permafrost soils and lake sediments (dark brown). For the arrows, a blue fill is used for fluxes associated with water (dissolved and suspended), while the grey fill is used for fluxes related to the transport of particles. The background image is looking south over the TBL catchment with the Greenland ice sheet to the left.

divided between eolian material, an easily exchangeable fraction (leachable using ammonium acetate) and the difference between the total sediment accumulation and these two fractions. The latter is interpreted as representing either more stable authigenic precipitates or elements associated with organic material.

For the terrestrial system eolian deposition and erosion rates were assumed equal because: i) there are currently no signs of newly formed sand dunes or patches with buried vegetation (Petrone et al., 2016), ii) the thick eolian deposits that cover the catchment formed during earlier periods with higher eolian activity (Petrone et al., 2016; Rydberg et al., 2016; Willemse et al., 2003), and iii) even if there are deflation scars present on some ridges in the catchment they are not rapidly expanding (Heindel et al., 2015). This assumption means that Eqs. (2) and (3) can be simplified and rearranged as follows.

$$\text{Dep\_ter} - -\text{Dis\_ter} = \text{Acc\_ter} - -\text{Rel\_ter} \tag{3}$$

$$\begin{aligned}\text{Dis\_ter} + \text{Dep\_aq} + \text{Eol\_aq} - -\text{Exp\_tal} - -\text{Exp\_out} - -\text{AccSed\_eol} - \\ -\text{AccSed\_NH4} - -\text{AccSed\_res} = \text{Acc\_aq} - -\text{Rel\_aq}\end{aligned} \tag{4}$$

After this rearrangement the Eqs. (4) and (5) show measured or estimated terms on the left-hand side and the unmeasured balancing terms on the right-hand side of each equation. If the balancing term is positive, it indicates that there is a net accumulation of the element in that system, while it indicates a net release if it is negative.

2.3. Chemical sampling and data set

Full details on sampling and analysis, as well as, a very brief overview of the data collected until 2015 can be found in an Earth Systems Science Data-paper, (Lindborg et al., 2016). Accompanying Lindborg et al. (2016) is also a PANGAEA-data base (https://doi.pangaea.de/10.1594/PANGAEA.860961 March 2023), which has been updated to include all data from the TBL-site collected until 2019. Average, minimum and maximum concentrations, as well as, standard deviations (1σ) for each sample type can be found in the supporting information (SI-Table 2). Below follows a short description of the data used in the mass-balance budget calculations together with an explanation of the assumptions that have been made and why some data points were excluded.

To represent wet deposition, rain-water samples collected during the period 2011–2018 were used (n = 8). Two rain samples from August 2011 were excluded due to likely contamination by bird droppings that, e.g., gave phosphorus values 400 times higher than uncontaminated samples. Surface runoff during the snowmelt season was based on filtered snow samples collected in old snowpacks during the period 2011–2018 (n = 6; two samples were excluded due to extremely high calcium values) and water collected in small temporary streams during the snowmelt season (n = 13). For the sub-surface flow through the active layer during summer and autumn the average of all groundwater chemistry-data from lysimeters (n = 65) and soil–water wells (n = 26) between 2011 and 2018 were used. The lake-water chemistry was represented by the average of all filtered lake water samples collected from 0.5-, 3-, 5- and 25-m water depth at the center of the lake during the period 2011–2018 (n = 13). Elemental concentrations in all water samples were analyzed by inductive coupled plasma, sector-field mass-spectroscopy (ICP-SFMS) at ALS Laboratories in Luleå, Sweden.

For elements with a limited number of values below the reporting limit, we replaced below reporting limit values with half the reporting limit. This procedure will might over- or underestimate the average values used in the mass-balance budget, but using half the reporting limit tends to have a less drastic effect on the average as compared to replacing with zero or the reporting limit (Helsel, 2006). Another option would be to remove these elements completely, but in this case, we opted to retain as large a number of elements as possible. For all elements except Nb (where all lake-water samples were below the reporting limit), all catchment pools had at least one sample that was above the reporting limit, which indicates that using half the reporting limit should give an average of that is of the correct magnitude. For Cr prior to 2012, Sc prior to 2018 and Sn prior to 2013 data below the reporting limit during the earlier part of the study period had to be removed entirely because an increased analytical sensitivity meant that the reporting limit dropped by at least one order of magnitude between the earlier and latter part of the study period.

The biomass of terrestrial vegetation and aquatic macrophytes were determined through destructive sampling of sample plots from all vegetation types (Lindbort et al. 2016 and 2020). All available solid-phase chemical data for both terrestrial plants and litter (n = 7), and aquatic macrophytes (n = 7) were used. To remove the influence of eolian material – which is always found attached to the plant and litter samples – the elemental concentrations in terrestrial samples were corrected using the zirconium-to-element ratio in a sample of eolian material and the Zr concentration(assuming a negligible plant uptake of Zr in above ground biomass; Shahid et al., 2013). All available solid-phase chemistry data for quaternary deposits from active layer (n = 26) and permafrost (n = 11) were used (Lindborg et al., 2016). The chemical composition of eolian material was determined from material that had accumulated on vegetation in the TBL-catchment during periods with extensive eolian transport (n = 1). To ensure that this single sample was representative it was compared to samples from the glacial outwash plain (Sandflugtdalen) – which is located ~ 6 km southwest of the TBL catchment – and for the 23 elements analyzed in both samples the elemental compositions were similar (Fig. 3b; SI-Table 2). All available data on concentrations in suspended material (>0.45 μm) in the lake water – which includes phytoplankton and zooplankton – were used (n = 2). For the lake sediment, concentration data from one core collected in 2011 and all cores collected in 2012 (n = 13) were used.

Total elemental concentrations in all solid-phase samples, except for the sediment cores collected in 2012, were analyzed by ICP-SFMS following either lithium borate fusion, strong acid digestion or sintering depending on the specific element (Lindborg et al., 2016). In addition to total concentrations, the easily exchangeable fraction was analyzed by ICP-SFMS following leaching in ammonium acetate (NH_4Ac) in a number of soil-phase samples (n = 25) and the lake sediment samples collected in 2011. The lake sediment samples collected in 2012 were analyzed for total concentrations using wave-length dispersive X-ray fluorescence spectroscopy (WD-XRF) following the procedure described by Rydberg (2014). To evaluate similarities and differences between samples from different compartments and identify potential outliers, all water and solid-phase samples were subjected to a principal component analysis (PCA). Prior to PCA elements where the majority of observations were below the reporting limit were removed (resulting in 37 and 23 elements for the water and solid phase, respectively), and the data was converted to z-scores (average = 0; variance = 1) to remove any effects of scaling. All PCs with Eigen values above one were extracted, and a Varimax rotated solution was used.

2.4. Calculations of elemental transport fluxes

Input to the catchment as wet deposition, on both the terrestrial and the aquatic systems, was calculated based on the average concentration in rainwater, the annual precipitation (269 mm), and the terrestrial (1.7 km^2) and lake area (0.37 km^2), respectively (SI-Table 3). Because of the eolian activity, dry deposition was assumed to be predominately related to eolian deposition. As discussed above we also assumed that eolian deposition and erosion were equal in the terrestrial system. For the aquatic system the estimate of eolian deposition was based on the

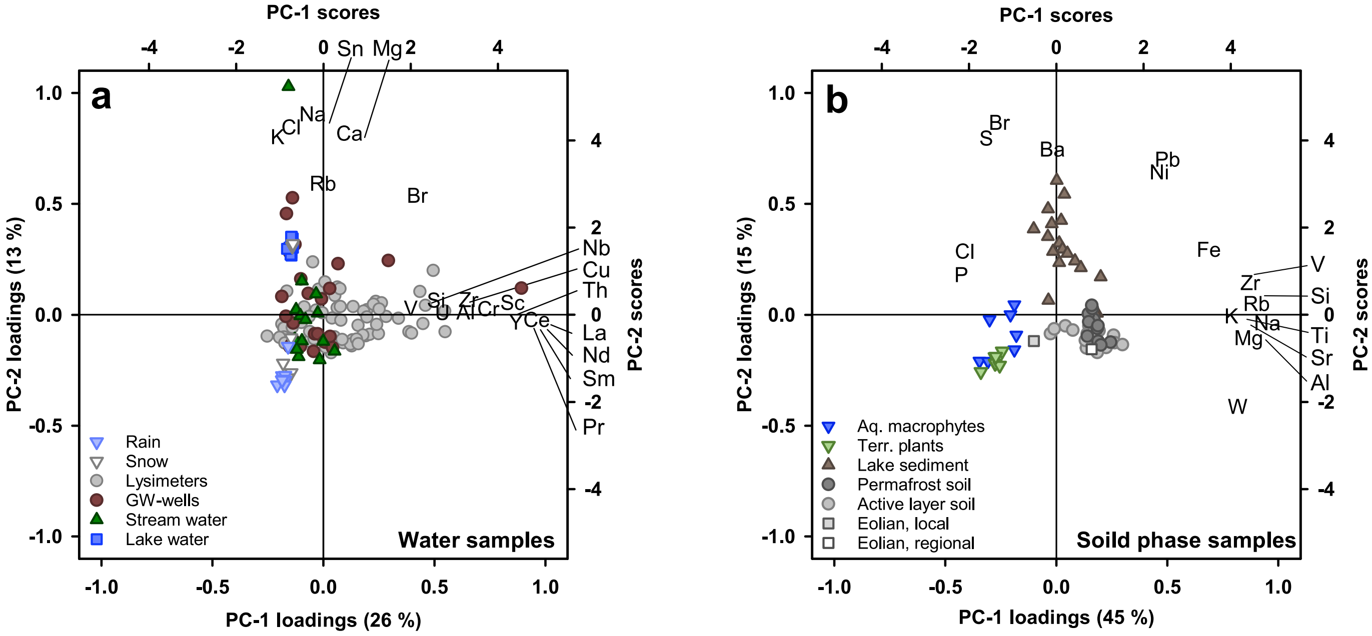


Fig. 3. Combined Loadings/Scores plots for the two first principal components (PC-1 to PC-2) of the PCA for the water (a) and solid-phase (b) samples, respectively. To increase the readability the lables for some elements are moved to the side of the plot and their locations in the loadings plot are represented by the solid black lines.

assumption that Zr mainly is associated with mineral particles in the coarse silt and sand fractions (Hood et al., 2019; Taboada et al., 2006), and that the lack of larger streams means that such large mineral particles only enter the lake as a result of eolian deposition (this assumption is supported by Zr concentrations less than 1 µg/L in most water samples). Based on extremely clear water (Secchi depth more than 15 m) and the very small amount of suspended lithogenic elements in the lake water (e.g., 0.001 µg Zr $L^{-}1$ and 0.8 µg Al L^{-1}) it is reasonable to assume that the relatively large silt and sand-sized particles that enters the lake through eolian deposition sink through the water column rapidly. This allows us to use the element-to-zirconium ratio in eolian material together with the sediment accumulation rate for each element to estimate the eolian deposition to the lake surface (and the eolian fraction of the lake sediment pool). For some elements the estimated eolian input was larger than the total amount found in the lake sediment. In such cases the eolian deposition and eolian fraction was assumed to be equal to the total concentration minus the easily exchangeable fraction (from NH_4Ac-leaching).

According to the hydrological modelling (Johansson et al., 2015b), about 52% of the annual runoff from the terrestrial catchment to the lake occurs via small temporary streams and the uppermost active layer during the snowmelt season, whereas the remaining 48% of the annual runoff occurs as a diffuse sub-surface flow during summer and fall. The hydrological transport of elements to the lake during the snowmelt season – when the active layer is shallow and the water has limited contact with the soil matrix – was based on the chemical composition in samples collected in temporary streams and the flow of water from the entire catchment (1.7 km^2; SI-Table 3). For remainder of the year – when the active layer is thicker and the water has been in contact with the soil matrix for an extended time – we used the chemical composition in water from lysimeters and soil–water wells and only runoff from the originally defined catchment area (1.2 km^2) was assumed to reach the lake (SI-Table 3). The export of elements through the talik and the lake outlet was calculated from the average lake-water concentration and the flow of water through the talik and outlet, respectively (SI-Table 3). For the talik, only filtered lake-water samples were used, whereas for the export through the lake outlet, the calculations were based on both filtered lake-water and suspended material.

The annual sedimentation rate for the 22 elements analyzed using wave-length dispersive X-ray fluorescence spectroscopy (WD-XRF) was determined based on elemental concentrations and sediment bulk density from 13 sediment cores collected across the lake basin in 2012. Sediments are known to undergo post-sedimentation diagenetic changes that might release easily degraded material back to the water column (Gälman et al., 2008; Rydberg and Martinez-Cortizas, 2014) and several of the sediment profiles included subsurface peaks in Fe and Mn, which likely are the result of diagenetic redox processes (Davison, 1993). To reduce the effect of such diagenetic processes – and to calculate the net sedimentation rate over time – the elemental concentrations for 0–3 cm samples were replaced by the average concentration for 3–8 cm (but using the bulk density from the 0–3 cm samples). For the sediment core collected at 17 m water depth, i.e., in the transition zone between oxygenated and anoxic water (Pangaea ID 049; Lindborg et al., 2016), the Fe-peak was located at 4–5 cm sediment depth, and hence, the concentrations for the 0–5 cm section were replaced by the average for the 5–10 cm section for this core.

We then calculated the total inventory in each core for the last 150-year period (based on Pb-210 and C-14 analyses; Lindborg et al., 2016), which was then used to calculate the accumulation for the entire lake basin using the same approach as used in Lindborg et al. (2020). In brief, the lake basin was divided into sub-units either based on bottom type (shallow erosion bottoms, vegetated areas and deep accumulation bottoms) or on similarities in the sediment composition (depth and location). The sediment accumulation for each sub-unit of the basin was then based on the areal extent and the average of all sediment cores for that sub-unit. This sediment accumulation rate was then recalculated to an annual sediment accumulation rate for the entire lake basin by summing all sub-units and dividing by 150 years. The mass-balance model then used the average of these two approaches to represent the sediment accumulation rate for each element. In order to estimate a sediment accumulation rate also for the 20 elements not analyzed in the 13 cores from 2012 (analyzed using WD-XRF) we first calculated the sediment accumulation rate (kg yr^{-1}) for the entire basin using the annual sedimentation rate for the 22 elements in the 13 sediment cores from 2012. The annual sedimentation rate was then used together with the elemental concentrations for the additional 20 elements from the sediment core collected in 2011 that had been analyzed using ICP-SFMS (Pangea ID 035; SI-Table 3 and 4). For all 42 elements the estimates of the easily exchangeable fraction were based on the ratio between the exchangeable and total concentrations for each respective element in the sediment core from 2011 (Pangea ID 035).

2.5. *Calculations of pools*

To calculate the elemental pool in terrestrial above-ground biota the average elemental concentrations of all plant and litter samples were multiplied by the area specific estimates for above ground biomass including litter (weighted averages based on vegetation types; (Lindborg et al., 2020; Lindborg et al., 2016; Petrone et al., 2016) and the entire terrestrial catchment area (excluding exposed bedrock; SI-Table 3). Similarly, for the aquatic biota the average chemical concentrations in aquatic macrophytes were multiplied by the estimated macrophyte biomass from Lindborg et al. (2020; no weighting was done between different vegetation zones; SI-Table 3). The aquatic-biota pool only includes the aquatic macrophytes, because the other pools were either too small to be relevant (i.e., benthic fauna) or were included in the lake-water suspended matter pool (i.e., various types of plankton; Lindborg et al., 2020). The active-layer groundwater pool was estimated from the average chemical composition in samples from lysimeters and soil–-water wells, the entire catchment area (excluding exposed bedrock), the average groundwater level (Johansson et al., 2015a; Johansson et al., 2015b), the maximum active layer depth (weighted average based on vegetation types; (Petrone et al., 2016)), and the average active layer soil porosity (excluding 0–10 cm; SI-Table 3). The lake-water pool was derived from the lake volume and the average concentrations in both filtered samples (dissolved fraction) and on filters (suspended material; SI-Table 3). Because of this the lake-water suspended-matter pool also include phyto-, bacterio- and zooplankton larger than 0.45 µm.

The terrestrial solid-phase soil pool was divided into an upper active layer and a deeper permafrost pool (the latter includes also frozen ground water; SI-Table 3). Because the aim of this study primarily was to assess the present-day situation, we have excluded the bedrock and deeper quaternary deposits (below 1 m), which have limited exchange with other catchment pools. For the active layer, the total pool was estimated from concentration data for all soil samples collected within the active layer, the maximum active layer depth (weighted average based on vegetation types; Petrone et al., 2016), the size of the entire catchment area (excluding exposed bedrock) and the soil bulk density (SI-Table 3). Because several of the soil samples were analyzed for both total concentrations and an easily extractable fraction (NH_4Ac-leaching), the active-layer pool was divided into a recalcitrant and an easily exchangeable fraction (e.g., elements adsorbed to mineral particles or organic material).

The particulate phase of the permafrost pool was estimated from the chemical concentrations in long soil cores collected in 2014 and 2016, the volume below the active layer (down to 1 m), the size of the entire catchment area (excluding exposed bedrock) and the same bulk density as for the active layer (SI-Table 3). For the frozen ground water, the calculations were based on the same data as for the active layer, but using the soil volume below the permafrost table. Because the frozen groundwater is immobile, the solid-phase and groundwater pools were combined into a single permafrost pool. No direct measurements of the sediment volume have been made in TBL, and therefore the sediment pool was calculated from the sediment accumulation rate and the estimated minimum age of the lake (i.e., 4000 years; Lindborg et al., 2020; SI-Table 3). In a similar way as for the active-layer pool, the sediment pools could be separated into three fractions, i.e., an eolian fraction, an easily exchangeable fraction and a residual fraction. The residual fraction is interpreted as either relatively stable authigenic precipitates or organic complexes that were not dissolved by the NH_4Ac-leaching.

3. Results

3.1. Principal component analysis and validity of the data

The water-sample PCA identified eight principal components (with eigenvalues above one), together explaining 84% of the variance in the dataset (Fig. 3a). Principal component 1 (PC-1) is related to high concentrations of many tri- and tetra valent metals with low solubility (e.g., Nd, La, Pr, Sc, Th, Y, Zr; (Deer et al., 1992)). On the negative side of PC-1, we find precipitation and lake-water samples, which tend to have low concentrations of these elements, whereas the surface runoff and lysimeter samples range from positive to negative scores. PC-2 separates lake-water samples on the positive side from precipitation on the negative side (with surface runoff and lysimeter samples in the middle). This PC is related to relatively mobile, water soluble, elements that are supplied by wet deposition and that are enriched in the lake water due to evaporation (e.g., Cl, Na, Sr, Mg, K, Ca; Johnson et al., 2000). The remaining six PCs are driven primarily by a small number of groundwater samples and they are difficult to relate directly to any specific process. The fact that a small number of samples drive a large part of the remaining variation could indicate that these samples are not representative and should be removed. However, even when the strongest outliers are removed from the PCA, a similar pattern appears. Furthermore, temporal variability in water chemistry can be significant in Arctic environments (Pokrovsky et al., 2012), and it is likely that these samples do reflect true variability. Hence, all available water chemistry data was used when calculating the averages used in the mass-balance budget.

Rain and fresh snow samples exhibit very little temporal variability (Fig. 3a), which suggests that even a relatively low number of samples collected in different years should give a representative average for the atmospheric deposition, and that no weighting between seasons or years is necessary. Similarly, TBL lake water samples are also consistent over time, and should give a representative average for the lake water pool. Although there is more variability in the soil-water, lysimeter and stream-water samples, there is considerable overlap (Fig. 3a). That stream and soil–water samples tend to have slightly negative PC-1 scores, while lysimeter samples range from slightly negative to positive scores, can likely be attributed to differences in the contact time between the water and the soil particles. During the snowmelt period, when most stream-water samples were collected, the contact between the water and soil particles is limited due to the frozen active layer. When the active layer has thawed, and lysimeter samples can be collected, the contact increases, resulting in higher concentrations of elements released due to weathering. In the mass-balance budget, this difference in hydrochemistry is handled by separating the runoff into a snowmelt and non-snowmelt component. Apart from the differences between the snowmelt and non-snowmelt seasons, there are no clear systematic patterns in the chemical composition between years or seasons. Hence, we have not done any weighting of the chemistry data for the non-snowmelt period.

For the solid-sample PCA there was even higher consistency among sample types, and only four PCs were extracted (explaining a total of 78% of the variance; Fig. 3b). That the sample types group closely together indicates that the average should be representative for the chemical composition of that pool. PC-1 separates mineral soils from biotic samples, with elements associated with mineral particles (e.g., Zr, V, Si, Al, Rb, Ti, K, Na, Mg, Sr) as the main drivers. PC-2 is related to S, Br, Ba, Pb and Ni, and higher PC-2 scores are mainly associated with the sediment samples. Higher concentrations in these elements, which all can be associated with organic matter, might be related to the enrichment of these elements in the older and more degraded organic material found in the sediment samples (Biester et al., 2004). PC-3 is driven by a small number of sediment samples that are rich in Mn and Fe, likely because of diagenetic enrichment due to redox processes within the sediment (Davison, 1993). Because such enrichment does not represent a net accumulation in the sediment, these samples were not included in the sedimentation rate calculations (see method section). PC-4 is driven by small variations in Ca, with samples with higher concentrations plotting on the positive side.

3.2. Fluxes of elements

Based on similarities in their chemical properties and behavior, we have assigned the 42 elements to eight groups (Fig. 4; Table 1). Zirconium (Zr), aluminium (Al), titanium (Ti), scandium (Sc), silicon (Si), niobium (Nb), vanadium (V) and yttrium (Y) are assigned to the *Particle bound elements*-group. This group consists of elements where the main source is eolian deposition, and that tend to be associated with silicate minerals and that generally have a low solubility. The input as wet deposition is virtually non-existent, and the release from the terrestrial system is generally low. For all elements in this group the main sink is the lake sediment (eolian fraction), but the mass-balance budget also suggests a small accumulation of Sc, Si, Nb, Y and V in the lake water.

In the second group – *Lanthanides/Actinides* – we find lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), samarium (Sm), thorium (Th) and uranium (U). Also for this group the main source is eolian deposition, but there is also a substantial internal release from the terrestrial system. The main sink is the sediment, but even if the eolian fraction dominates, a considerable part of the sediment pool is found in the exchangeable and residual fractions. For Th the release from terrestrial soils is not enough to close the mass-balance budget, which appears as a source in the aquatic system (Rel_aq). The mass-balance budget suggests a minor accumulation of Sm, Nd and Pr, as well as, a considerable accumulation of U in the lake water.

The third group – the *Alkali/Alkaine earth metals* – consists of lithium (Li), sodium (Na), potassium (K), rubidium (Rb), magnesium (Mg), calcium (Ca), strontium (Sr) and Barium (Ba). Similar to the two previous groups eolian deposition makes an important contribution to the input, but wet deposition is also a prominent source. For Li, Ca and Mg there is also a considerable release from the terrestrial system, while K is retained in the terrestrial system. A notable difference from the previous groups is that, except for Ba, there is a substantial accumulation of these elements the lake water, and that the sediment is a less important sink. The fact that the Alkali and Alkaline earth metals tend to accumulate in the lake water also leads to an export of these elements through the outlet and the talik.

Iron (Fe) and manganese (Mn) form the *Fe/Mn*-group. Similar to the Particle bound elements and the Lanthanides and Actinides, the main source for Fe and Mn is eolian deposition, and they accumulate in the lake sediment. For Fe, the sediment accumulation is substantially higher than the input to the aquatic system, which indicates that there is an internal source in the aquatic system (Rel_aq).

The *Metals/Metalloids*-group contain chromium (Cr), cobalt (Co), nickel (Ni), molybdenum (Mo), copper (Cu), bismuth (Bi), tin (Sn), cadmium (Cd), zinc (Zn), and antimony (Sb). Similarly to the Alkali and Alkaline earth metals, the main source for most of these elements is atmospheric wet deposition, but for Co, Ni and Mo there is also a substantial input from eolian deposition and release from the terrestrial system. For Sn, Bi, Cd, Zn and Sb 30–75% of the input is retained in the terrestrial system, while the aquatic system is the main sink for Cr, Co, Ni, Mo and Cu.

The *Halogen/Boron*-group contain boron (B), chlorine (Cl), bromine

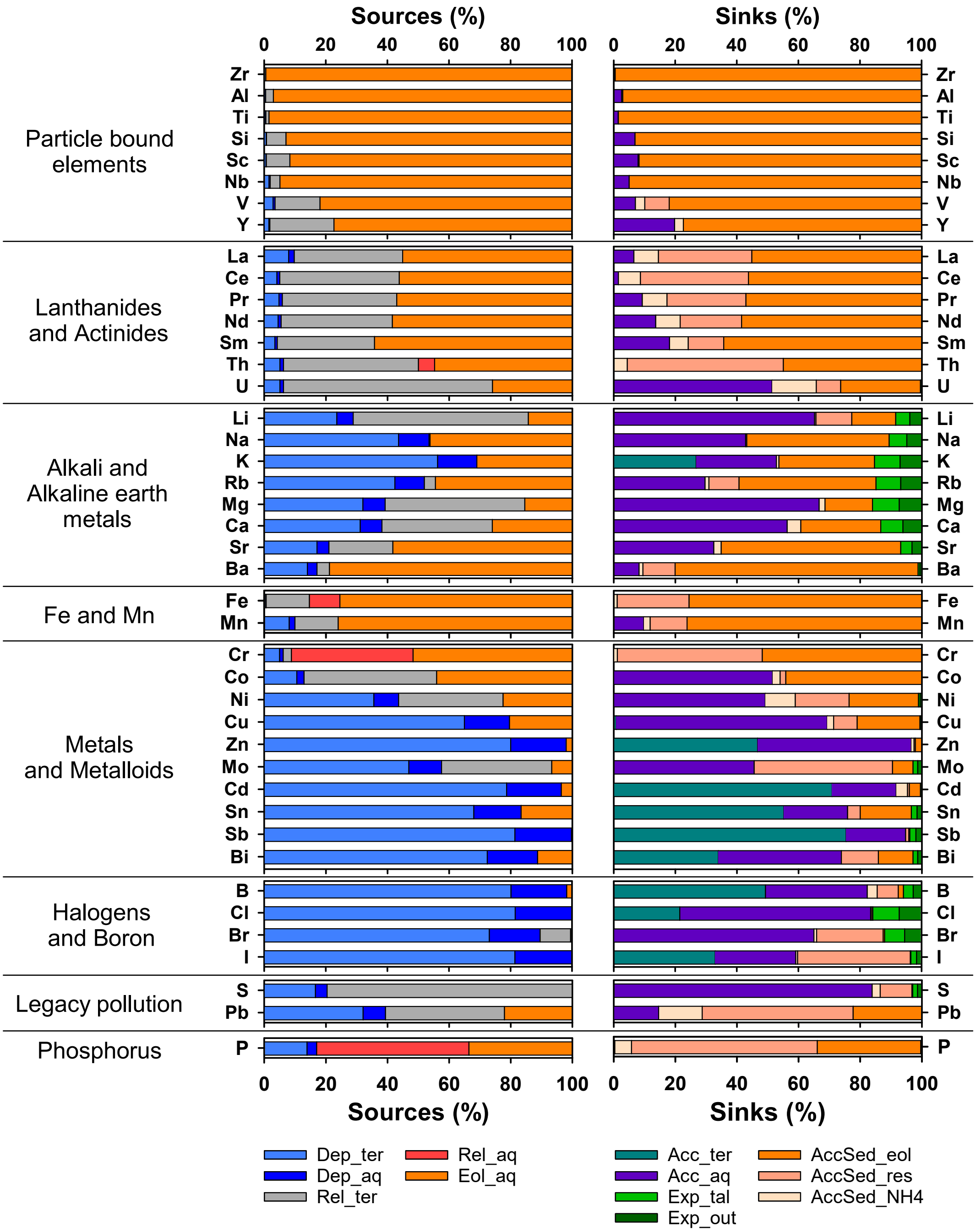


Fig. 4. Comparisons of element fluxes in the TBL catchment, i.e. for the combined terrestrial and aquatic systems. Primary sources are: Atmospheric deposition to the terrestrial (Dep_ter) and aquatic systems (Dep_acc), and eolian deposition to the aquatic system (Eol_lake). Sinks are represented by: Accumulation in terrestrial soils (Acc_ter), lake water (Acc_lakew), and the Lake sediment, which is divided into an Eolian fraction (AccSed_eol), a residual fraction (AccSed_res) and an Exchangeable fraction (AccSed_NH4). Internal sources are either terrestrial weathering (Rel_ter) or an internal source in the aquatic system (Rel_lakew).

Table 1
Grouping of the 42 elements into 8 elemental groups based on their respective behavior in the mass-balance budget.

Element group	Elements
Particle bound elements	Zr, Al, Ti, Si, Sc, Nb, V, Y
Lanthanides and Actinides	La, Ce, Pr, Nd, Sm, Th, U
Alkali and Alkaline earth metals	Li, Na, K, Rb, Mg, Ca, Sr, Ba
Fe and Mn	Fe, Mn
Metals and Metalloids	Cr, Co, Ni, Cu, Zn, Mo, Cd, Sn, Sb, Bi
Halogens and Boron	B, Bl, Br, I
Legacy pollution	S, Pb
Phosphorus	P

(Br) and iodine (I), which enter the system almost exclusively through atmospheric deposition. Three of the elements, B, I and Cl, are retained in the terrestrial system, while Br shows a small release. All Cl that enters the aquatic system accumulates in the lake water, but for the other three elements there is also an accumulation in the exchangeable and residual fractions of the sediment with a higher accumulation of I than for Br and B. That the Halogen/Boron-group tend to accumulate in the lake water also leads to an export through the outlet and talik,

Sulphur (S) and lead (Pb) are grouped together because they are both known *Legacy pollutants*. Both of these elements are supplied through release from the terrestrial system. Sulphur is mostly retained in the lake water, while Pb accumulates in the lake sediment. *Phosphorus* (P), that enters the system as both wet and eolian deposition, form a group of its own. The main source to the system is an unidentified source in the aquatic system (Rel_aq), and the main sink is the sediment, where most of the P is associated with the residual fraction (AccSed_res).

3.3. Catchment pools

When assessing how the elements are distributed across the different pools the general patterns are relatively similar between elements (Fig. 5). That is, the catchment pools that contain the largest mass of solid matter – i.e., soils and lake sediments – also tend to contain the largest portion of a specific element, while smaller pools – e.g., groundwater, lake water and biotic pools – generally only make up less than 1% of the total catchment pool. The fact that the physical size of a pool is the most important factor in determining the pool size also means that about 90–98% of the mass of most elements are found in the terrestrial system. When comparing the different fractions of the active-layer soil and lake sediment pools, the easily exchangeable fraction is in general one to two orders of magnitude lower than the more recalcitrant fractions. The most notable exceptions from this pattern are the halogens, where more than 30% of the catchment inventory is found in the aquatic system (Fig. 5). There are also a number of other elements that generally are soluble under oxidizing conditions (i.e., S, B, Sb and Mo) where more than 10% of the catchment pool is found in the aquatic system.

When looking into the details it also becomes evident that there are other differences between the elements. A larger part of the catchment pool of halogens, B, S, Sb and Zn are found in the groundwater pool as compared to other elements, and the biotic pools are enriched in, e.g., B, Cl, P, S, Mn, Mo and Zn (for the aquatic macrophytes also Br and I are enriched). Furthermore, a number of elements for which atmospheric deposition is the most important source have a larger proportion of the catchment pool in the active-layer pool relative to the permafrost pool. This is most evident for the halogens, but also S, Pb, Sb, K and B display this pattern. In the terrestrial system Cl stands out by having almost the entire active-layer soil pool in the exchangeable fraction, and in the lake sediment more than 70% of the sediment pool for B, Br, Cl, I, Mo, S, Sb and Pb is found in the non-eolian fraction (i.e., either in the exchangeable fraction, as authigenic precipitates or associated to organic complexes).

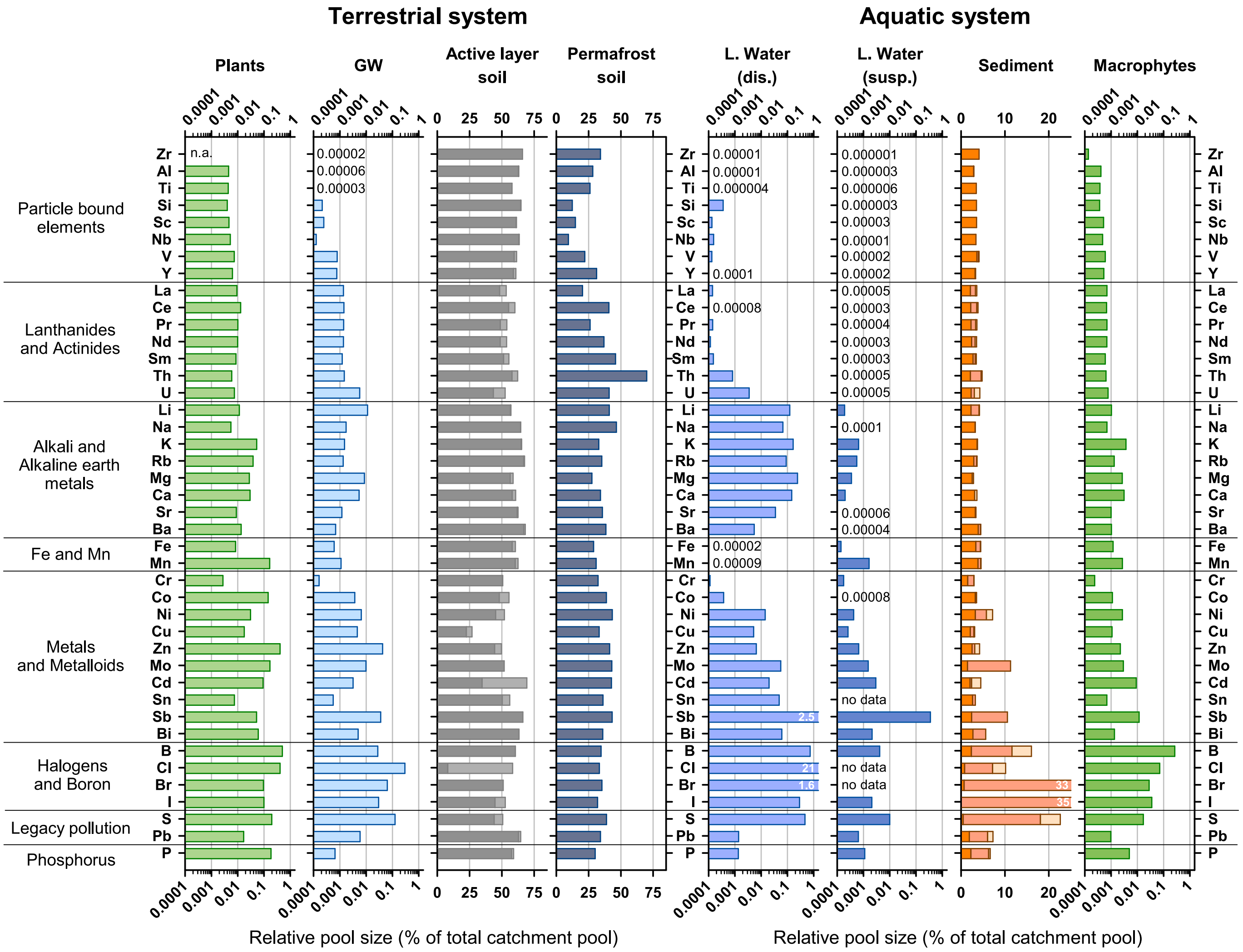


Fig. 5. Relative pool sizes for the 42 elements (calculated as the percentage of the total catchment pool) for terrestrial plants (Ter. plants), groundwater (GW), active-layer soils, permafrost soils, dissolved and suspended substances in lake water (L.water), lake sediment, and aquatic macrophytes (Aq. macrophytes). For the active-layer soil-pool the light and dark color represent the exchangeable and residual fractions, respectively. For the lake sediment pool the three fractions (eolian, residual and exchangeable) are represented by different shades of orange with a lighter shade representing a more easily mobilized fraction. To increase readability, pools representing less than 1% of the total catchment pool are plotted on a logarithmic scale, and pools representing less than 0.0001% are presented as numbers instead of bars. For the lake sediment the relative pool size for pools larger than 20% are given as white numbers on top of the bar.

4. Discussion

Our mass-balance model identifies eolian deposition as an important source for elements associated with silicate minerals (i.e., the Particle bound elements and Lanthanides/Actinides), which is consistent with previous studies in both the Kangerlussuaq region and other arid and semi-arid landscapes (Okin et al., 2006; Willemse et al., 2003). At the same time wet deposition is virtually the only source for the Halogen/Boron-group, and it makes up a considerable part of the input of many of the elements in the Alkali/Alkaline earth metals and Metals/Metalloids-groups (Fig. 4). The halogens (Br, Cl and I) can likely be traced back to the ocean, because the Cl:Br (251) and Cl:I (845) ratios in rain water collected at TBL are within the range that could be expected considering the distance and altitudinal difference compared to the sea (Duce et al., 1965; Willis et al., 2018). For the Alkali/Alkaline earth metals and Metals/Metalloids-groups the origin is more complex. Even if the Na:Cl (1.9) is similar to that found in the ocean, the higher K:Cl (0.50), Mg:Cl (0.40) and Ca:Cl (0.76) ratios in rain water collected at TBL as compared to the ocean indicate that there is an additional source. A possible explanation is a release of these elements from eolian dust trapped in the rain drops. For the Metals/Metalloids-group the origin can be both the ocean, release from eolian dust, and long-range transport of anthropogenic emissions (Heidam et al., 2004; Willis et al., 2018). Some of the elements in the Halogen/Boron and Metals/Metalloids-groups, as well as, K are retained in the terrestrial system. For K, this retention is likely at least partially related to uptake by biota (Peltola et al. 2008), while the retention of both the Halogen/Boron and Metals/Metalloids-groups likely is related to sorption and complexation with soil organic material, including the formation of organohalogens (Broder and Biester, 2015; Lidman et al., 2017; Svensson et al., 2017). In addition to being retained in the terrestrial system there is a considerable hydrological transport of the elements supplied with wet deposition to the lake. This hydrological transport is also important for all elements were the terrestrial system acts as an internal source through, e.g., weathering.

That the dissolved fraction of the lake-water pool is at least one order of magnitude larger than the suspended material pool (Fig. 5) indicates two things. First, that elements associated with mineral particles (e.g., the Particle bound element-group) that are deposited on the lake surface rapidly sink through the water column and accumulate in the sediment. Second, that most of the accumulation in the lake water occurs either as truly dissolved species, colloids or as soluble complexes with, e.g., organic material. A consequence of the dry climate is that TBL has a long water residence time and any elements entering the lake in dissolved form (e.g., Sr, Rb, Li, Na, Cl, K, Mg, Br) either tends to become enriched in the water column or form precipitates that accumulate in the sediment. Even if the mass-balance budget suggests that the concentrations of dissolved elements in lake water increase with time, TBL is still relatively dilute compared to some other lakes in the region (Anderson and Stedmon, 2007; Henkemans et al., 2018). This is likely a result of the lake being close to the icesheet, where precipitation is higher and the water-balance is less negative than 20–30 km towards the coast where most lakes with higher salinity are located.

4.1. *Weathering and soil development in a cold and dry landscape*

The cold and dry conditions in West Greenland suggest a relatively low weathering rate for mineral particles (Dixon et al., 2016). At the same time soils in areas close to the inland icesheet are young and undeveloped, which presumably leads to a faster weathering rate (Deuerling et al., 2019). This raises the question regarding what factor that is more important in the TBL catchment. For the Particle bound element-group the chemical composition of the lake sediment suggests that the transport is almost exclusively related to eolian processes. However, for the Lanthanides/Actinides and Fe/Mn-groups, which are known to be associated with relatively easily weathered minerals, e.g., apatite, pyroxenes and amphiboles (Land et al., 1999), the mass-balance budget indicates that there is a substantial release from the terrestrial system. There is also a substantial accumulation of authigenic precipitates or large, non-soluble, organic complexes in the sediment (i.e., SedAcc_res and SedAcc_NH4). This behaviour is consistent with a transport of dissolved weathering products from the terrestrial system to the lake, which shows that weathering is an important process also in this cold and dry climate.

In order to compare the extent of weathering in the TBL-catchment to warmer climates two of the lanthanides (La and Ce) can be used. The average solid-phase concentrations in TBL-soils are comparable to values reported for the upper continental crust and granitic bedrock in Europe (28 and 61 mg kg^{-1} for La and Ce, respectively; Minarik et al., 1998; Rudnick and Fountain, 1995; Stille et al., 2009). This implies that at least the total pool of La and Ce in the soil at TBL is similar to other sites with a similar bedrock. If we then look at the soil–water concentrations from lysimeters in the TBL-catchment (4 and 10 μg/L for La and Ce, respectively) they are an order of magnitude higher than in soil–-water samples in more weathered granitic upland soils in northeastern France (0.1–0.4 and 0.04–0.7 μg/L for La and Ce, respectively; Stille et al., 2009). Because lanthanides are relatively immobile unless they become associated with dissolved organic matter (Robinson et al., 2018), at least part of this difference can be attributed to differences in soil type. All soil–water samples from TBL are collected in the peaty soils in wetland areas, which are rather different from the upland soils studied by Stille et al. (2009). If we instead compare the TBL-samples to soil water from riparian soils in northern Sweden (granitic bedrock; 1.4–2.2 and 3.2–5.1 μg/L for La and Ce, respectively; Lidman et al., 2019), the concentrations are more comparable, but the TBL-samples are still higher. Even if the La and Ce concentrations in soil water also are affected by other factors than weathering rates (e.g., hydrological flow paths and contact time), the comparatively high La and Ce concentrations in soil water in the TBL catchment indicates that weathering is an important process, and that the cold and dry conditions are counteracted by the presence of easily weathered minerals in the terrestrial soils.

4.2. *Retention and transport of elements supplied by wet deposition*

The dry conditions and high evapotranspiration in the TBL-catchment imply a limited hydrological transport (Johansson et al., 2015b), but the mass-balance budget still identifies hydrological transport as being important for many elements (e.g., Lanthanides/Actinides, Alkali/Alkaline earth metals and Fe/Mn-group, as well as, for S and Pb). For Cl – for which wet deposition is virtually the only source – there is a tendency that it accumulates both in the terrestrial and aquatic system. That an element like Cl, which is generally considered to be conservative and mobile (Lockwood et al., 1995), tend to accumulate in terrestrial soils can help us to better understand the hydrological transport in this landscape.

Looking at the terrestrial pool sizes, 73% of Cl in the terrestrial system is found in the exchangeable fraction of the active layer, whereas only 14% is found in the deeper permafrost soils. The retention of Cl in the active layer, presumably as organically bound Cl (Svensson et al., 2017), is also evident from the Cl:Br-ratios. Stream water collected during the snowmelt period, which have had a limited contact with soil particles, have Cl:Br-ratios close to the precipitation (227 and 254 for stream water and precipitation, respectively), whereas water from deeper soil layers collected later in the season have Cl:Br-ratios that are much lower (i.e., 81 and 100 for groundwater wells and lysimeters, respectively). At the same time as there is a net retention of Cl in terrestrial soils, about 74% of the Cl that enters the terrestrial system through wet deposition is transported with runoff to the lake. One possible explanation for a simultaneous retention and release is that the transport with runoff is highly dependent on the hydrological connectivity between the different parts of the terrestrial system and the lake

(Moatar et al., 2017; Tiwari et al., 2017).

In the TBL-catchment this hydrological connectivity varies considerably between high and low flow situations, with the clearest example being that the north-eastern part of the catchment only is hydrologically coupled to the lake during the snowmelt period. Considering that the balance between precipitation and evapotranspiration is highly negative for most part of the summer period (Johansson et al., 2015b), it is reasonable to assume that also other parts of the catchment are decoupled from the lake for extended periods. If an area only produces runoff during the period when the active layer is frozen, this implies a limited movement of water through the soil and, in turn, a potential for retention also of relatively mobile elements like Cl. Dry and relatively warm conditions during the summer might also promote the formation of volatile organohalogens that can be emitted back to the atmosphere (which would show up in the mass-balance budget as a retention in the terrestrial system; Albers et al., 2017).

4.3. Release of legacy pollutants

In addition to the elements associated with weathering of mineral particles the mass-balance budget identifies the terrestrial system as a source for S and Pb. Although both are found in the local bedrock and potentially could be released through weathering (Auqué et al., 2019), they are also well-known pollution elements, and human emissions have caused an enrichment in soils also in remote areas (Hole et al., 2009; Renberg et al., 1994). Both S:Zr- and Pb:Zr-ratios in eolian material (0.38 and 0.011, for S:Zr and Pb:Zr, respectively) are lower than in soil samples from the active layer (2.5 and 0.023, för S:Zr and Pb:Zr, respectively). This implies that the active layer-soils are enriched in S and Pb compared to the original mineral material (the upper one meter of the soil consits of eolian deposits), which is visible as subsurface peaks in both S and Pb in a soil profile from one of the wetland areas in the TBL (SI-Fig. 1). Because wet deposition only makes up 17 and 47 % of the S and Pb that is transported from the terrestrial system to the aquatic system each year, this enrichment cannot be explained by the present-day wet deposition. This suggests that the TBL-catchment is in a non-steady state situation with regards to S and Pb, and fits well with the pollution history for these elements in West Greenland (Bindler et al., 2001; Hole et al., 2009). When the pollution load decreased in the 1980′s and 1990′s the terrestrial system started to act as a source for these legacy pollutants, something that has been reported also for more polluted areas closer to industrial activities (Giesler et al., 1996; Klaminder et al., 2010; Rydberg et al., 2010).

4.4. The lake-water column as a source for elements

For Fe, Cr, P and Th the mass-balance budget identifies the lakewater as an internal source (Rel_aq). Even if the exchangeable and residual fractions of the sediment shows that there is a considerable formation of, e.g., authigenic Fe-precipitates in the water column, the lake-water pool is not large enough to account for the rate of sedimentation. If we take Fe as an example, the annual sedimentation that is not related to eolian material (AccSed_res and AccSed_NH4) is about twice as high (1.7 times) as the entire lake-water pool (dissolved plus suspended material). A more realistic explanation for Fe would be that the apparent release is related to post-depositional redistribution of elements within the sediment (Davison, 1993). From the sub-surface peaks in Fe and Mn in some of the sediment cores we know that there is some redistribution of these elements in the sediment, which could lead to the sedimentation rate (and sediment pool) being overestimated. In order to circumvent this issue, we have not used the concentrations in the uppermost sediment samples most affected by redistribution in our calculations (see methods for further description). This means that our sediment accumulation rate should capture the long-term net accumulation, but there is still a risk that we do overestimate the net sediment accumulation rate.

Another possible explanation for the “missing” amount of these elements is that we either have an unidentified source or that the imbalance is related to uncertainties in our measurements and calculations. For Cr and Th the concentrations in both solid-phase and water samples are low, and the estimates for the sediment pool are based on measurements from a single sediment core that are extrapolated to the entire lake basin. Together this suggest that the imbalance is likely to be caused by uncertainties. For Fe the concentrations are higher and the estimates builds on a larger number of sediment cores, which makes it less likely to be related only to measurement errors. One possible explanation is that the water samples were not analyzed immediately after sampling (they were frozen and transported to the laboratory in Sweden). It is therefore possible that dissolved Fe^{2+} present in these water samples precipitated as Fe^{3+} prior to analysis (and thus filtered away prior to the analysis). This would lead to an underestimation of the hydrological input of both Fe and any elements that can co-precipitate with iron oxyhydroxides (e.g., Cr and P; Baken et al., 2015)).

For P it is also possible that geese (and other waterfowl) provide a direct input to the lake and near-shore area that our sampling does not capture (Dessborn et al., 2016; Van Geest et al., 2007). The Kangerlussuaq area is known as one of the main spring staging areas of *Anser albifrons flavirostris*, and TBL is visited by a large number of geese each year. According to the literature, the P content in geese droppings is around 5–6 mg g^{-1} dw., and the average dropping dry-weight is around 1 g for *Anser albifrons flavirostris* and 3.5 g for *Branta canadensis interior*, which are the two most common species in West Greenland; (Dessborn et al., 2016). Using the conservative 5 mg g^{-1} and 1-gram dry weight, it would be necessary to add 1.9 million droppings per year to the TBL lake or near-shore area to account for the 9550 g of missing P. Even if this sounds like an unrealistic number of droppings, it actually translates to only 11 droppings per square meter and year for the near-shore area of the lake (using a 50 m perimeter around the lake). From a survey in Svalbard the average number of droppings per square meter was found to be 8 (maximum 94; Jensen et al., 2019), and observations in the field at TBL (without actually counting) suggest that there are at least several droppings per square meter (both along the shore and in shallow waters). This suggests that at least a substantial part of the missing P could be related to birds and bird droppings, although further measurements would be needed to confirm this.

4.5. Uncertainties

When conducting catchment scale studies, the sheer size of the study object causes problems and it is virtually impossible to sample everything all the time, especially when studying a site in a remote location like West Greenland. In this particular case, this inherent weakness is to some extent counteracted by the use of the well-constrained, distributed, hydrological model developed by Johansson et al. (2015b). Their model uses 30 years of meteorological data from Kangerlussuaq together with site specific measurements to characterize the average hydrological situation for the TBL-catchment in terms of precipitation and hydrological flow. Based on their findings, our sampling effort could be focused on the areas and seasons with high hydrological flow (McClain et al. 2003; Laudon et al. 2004; Olefeldt and Roulet 2012). This introduces some bias in the sampling, but it also allows us to capture the most important aspects of the hydrological transport, both spatial and temporally (Johansson et al., 2015b; Umbanhowar et al., 2015; Wang et al., 2017).

One uncertainty in our mass-balance budget is the lack of direct measurements of eolian deposition and erosion. To solve this issue, we have assumed that eolian deposition and erosion cancel each other for the terrestrial system, and that we can use the element-to-zirconium ratio to estimate the eolian deposition on the lake surface. The first assumption can at least to some extent be justified based on aerial photographs, which clearly show that there is neither any significant build-up of eolian deposits nor any extensive growth of the existing deflation scares in the TBL-catchment over the last decades (Geodætisk institute; 1943, 1952, 1953, 1968 and 1985). For longer time scales, i.e., centuries to millennia, the fact that the catchment is covered by a one-meter-thick layer of eolian deposits clearly shows that there is a net input of eolian material to the TBL-catchment. For the use of the element-to-zirconium ratio one major caveat is that the ratio builds on a single sample, which raises the question as to how representative the used ratios are. Here we can draw confidence from the PCA (Fig. 3b) where the eolian sample and most mineral soil samples plot close together, indicating a similar chemical composition. At the same time, the concentrations of many trace elements are low and also small differences could have an effect on the mass-balance budget. Here there is an advantage of using a larger selection of elements, and focus more on how they behave as a group rather than looking at each individual element separately.

Accumulation in the lake water is another uncertain part of the mass-balance budget. First, it is a balancing term and all errors of the fluxes in the mass-balance budget are merged into this estimate. Second, the concentrations of many elements in the water samples are low, which further increase the relative importance of analytical errors for the budget. Because of the long residence time for both water and solutes in TBL a high accumulation of an element in the water column should be accompanied by a correspondingly sized lake-water pool. For most of the elements for which the mass-balance model suggests an accumulation in the lake water the lake-water pool is considerably larger (>30 times) than the annual input. In some cases, e.g., U, Y and Co, the lake-water pool is much smaller than the annual accumulation in the lake-water column (3, 0.5 and 0.4 times the annual input for U, Y and Co, respectively). Considering that the water residence time of TBL is 47 years, this suggests that the apparent accumulation in the lake water is mostly related to uncertainties in the data. For other elements, where the lake-water pool is smaller e.g., S (13) and Pb (6.5), this could be a result of the system not being in a steady-state. Finally, for Si (5.5) it is possible that the small accumulation in the lake water is associated with the incorporation of Si in diatom frustules (Meyer-Jacob et al., 2014). That this Si does not appear in the suspended matter pool suggests that epilithic and benthic diatoms growing on rocks and boulders, which are not included in our sampling, are an important part of the diatom pool.

Even if the use of multiple cores collected across the entire lake basin circumvents the issues with sediment focusing and ensures that we capture the spatial variability in the sediment accumulation (Blais and Kalff, 1995; Rydberg et al., 2012), it should be recognized that many of the trace elements (e.g., lanthanides and actinides) were only analyzed in a single sediment core. This means that for these elements we do not have any constraints on the spatial variability. Similarly, as for the eolian material discussed above, we can argue that the sediment composition is relatively constant at least for the 22 elements analyzed in all 13 cores (Fig. 3b). For the sediment pool it is also a potential problem that the thickness of the sediment has not been determined. This means that the total sediment pool was derived from the current sediment accumulation rate and the presumed age of the lake (4000 years), which introduces some uncertainty because we know that the sediment composition and sedimentation rate has changed with time (Rydberg et al., 2016). However, for the overall outcome of our study this is not a major issue, because the total sediment pool size is not used in any of the calculations in the mass-balance budget (for this purpose we use the accumulation over the last 150 years).

4.6. TBL in a changing climate

The prediction for the Arctic is that the future climate will be warmer with more precipitation, which will affect several aspects of the biogeochemical cycling at TBL (Boberg et al., 2018; IPCC, 2013). Previous studies have shown that the eolian transport is highly variable over time, and that it is affected by distance to the ice sheet, climate (wind and snow cover), vegetation and soil characteristics (Bullard and Austin, 2011; Heindel et al., 2017; Okin et al., 2006; Rydberg et al., 2016; Willemse et al., 2003). A warmer climate will eventually lead to a retreat of the ice sheet (Funder and Hansen, 1995), which initially might result in an increased eolian activity because “new”, un-vegetated, land will serve as source areas for eolian material (Okin et al., 2006). When the ice front moves even further inland, and the vegetation become denser, the eolian activity will eventually stop altogether in the TBL-catchment (Rydberg et al., 2016; Willemse et al., 2003). This reduced input of “fresh” eolian material will, with time, result in soils becoming more depleted in easily weathered minerals (Fritz et al., 2004; Klaminder et al., 2011). This will partly be counteracted by the warmer climate giving faster process rates, but considering that the present-day soil–water concentrations of La and Ce is actually higher in TBL as compared to in comparable soils from a site in northern Sweden (see discussion on weathering above), it is likely that the net effect will be a reduction in the weathering rate. The response may, however, vary for different elements depending on their weathering kinetics and what time scale that is considered.

Even if climate change potentially could alter the concentrations of various elements in rain and snow – e.g., through the influence of sea ice on the production of marine aerosols (Willis et al., 2018) – it is likely that a wetter climate would lead to an increased input of most elements supplied with wet deposition. Based on our mass-balance budget this would imply a larger retention of some elements (e.g., K, Cl, I, and several elements in the Metals/Metalloids-group) in the terrestrial system (Lidman et al., 2017; Lidman et al., 2014; Svensson et al., 2017). However, that the climate becomes wetter also suggests an increased hydrological transport of solutes and – if more permanent streams start to appear in the catchment – particles to the aquatic system (Wang et al., 2017). Here it should be noted that even if the climate becomes warmer with more precipitation, this doesn’t necessarily result in wetter conditions because increasing temperatures also lead to an increased evapotranspiration (Boberg et al., 2018).

A warmer and wetter climate also implies that several processes, e.g., primary production and organic matter decomposition, will become faster. A higher primary production implies that more organic matter is produced, but faster decomposition and melting permafrost could outweigh this effect and lead to a reduced storage of both organic matter and elements associated with organic matter (Berg, 2014; Olid et al., 2020). A higher decomposition rate could also lead to a higher production and transport of dissolved organic material (Laudon et al., 2012; Zhang et al., 2017), which would act as a transport vector for organically bound elements, e.g., weathering products (e.g., Lanthanides/Actinides) and legacy pollutants (e.g., S, Pb and Hg; Rydberg et al., 2010).

The hydrological transport is also affected by the presence of permafrost in the TBL-catchment. If the permafrost melts and the thickness of the active layer increases, water can flow along deeper flow paths. This will potentially mobilize elements that are currently stored in the permafrost, but it will also reduce the flow of water through the upper soil horizons that are rich in organic material and organically bound elements (MacLean et al., 1999). Whether an increased transport of dissolved elements to the lake will result in an increased enrichment in the lake water would largely be dependent on the water balance. Under present conditions, most of the water that enters TBL leaves through evaporation and the residence time in the lake for a conservative solute is 370 years (calculated as the lake volume divided by the outflow through the outlet and talik). If the lake level rises and outflow occurs more regularly there will be a much more pronounced export of solutes and particulate material to the down-stream river system, which would reduce the enrichment of solutes in the lake water.

5. Conclusion and the way forward

In response to the first of our research questions – i.e., how the combination of low temperatures and young, undeveloped, soils affect the release of weathering products – our mass-balance budget shows that weathering is an important source for elements found in more easily weathered minerals (e.g., Ce, La). Looking at the second question – i.e., the retention and transport of easily dissolvable elements – we can see that there is a considerable retention of several elements (e.g., Cl, I, B, K) in the terrestrial system. The fact that elements like Cl – which is normally considered to be relatively mobile – are retained suggest that there are parts of the catchment that have a poor hydrological connection with the aquatic system (i.e., that water leaves the soil through evapotranspiration rather than as runoff). However, because other parts of the catchments are hydrologically connected to the aquatic system, runoff still plays an important role for the transport of both weathering products (e.g., Ce and La) and elements found in wet deposition (e.g., Cl, Br, Li, Ca, Mg). As for our third question – i.e., are there any signs of release of legacy pollutants – there is a considerable release and hydrological transport of both S and Pb from the terrestrial system to the lake that cannot be explained by the current input. Together this highlights the need to integrate the terrestrial and aquatic systems when studying elemental cycling on a landscape scale, and we hope that our study can provide an increased understanding of biogeochemical cycling in Arctic landscapes. For future studies it would be preferable to have a more systematic sampling of precipitation and to, even if it is difficult, make an attempt to empirically quantify the net effect of eolian processes in the terrestrial system. Finally, we conclude that the many counteracting processes and feedback mechanisms makes it difficult to draw far-reaching conclusions regarding the effects of future climate change based on this study alone. One approach that might increase our ability to predict how future climate changes will affect the biogeochemical cycling in the Arctic would be to compare the relative importance between different fluxes and pools in catchments across climate gradients.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data is publicly available through the PANGEA-database (https://doi.pangaea.de/10.1594/PANGAEA.860961).

Acknowledgements

This work was conducted as a part of the Greenland Analogue Surface Project (GRASP) funded by the Swedish Nuclear Fuel and Waste Management Company (SKB). The authors would like to thank the Greenland Analogue Project (GAP) and Kangerlussuaq International Science Support (KISS) for providing logistical support throughout the years. We also thank two anonymous reviewers and the associate editor for their valuable help with this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.catena.2023.107311.

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