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A Lead (Pb) Mass-Balance Budget for a Dry Periglacial Catchment in West Greenland

-Discussing the fate of pollution Pb

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Abstract

Lead (Pb), occurs both naturally and as a pollutant in Arctic landscape systems. The ongoing climate change, especially pronounced in the Arctic, changes the premises for Pb transport and mobility. Thus, to predict future development of both natural and pollution Pb, an increased understanding of their storages and flows in an Arctic system is of interest. Here, a Pb mass-balance budget for an entire catchment in West Greenland was calculated. The budget shows that most Pb is stored in terrestrial soils (94%), while 6% of total catchment Pb is in lake sediments. Other Pb-pools are small in comparison (<1% combined). The entire catchment system has a negative balance, with annual inputs of 44 g Pb from precipitation and 67 g Pb from eolian deposition, while 150 g Pb is removed from the system (through sedimentation). Limited lake water outflow (0.15 g Pb yr^{-1}) suggests that the catchment in its entirety acts as a Pb-sink. The terrestrial system, however (wet deposition input of 34 g Pb and hydrological export of 68 g Pb annually), is considered to be a Pb-source to the aquatic system. The magnitude of hydrological and eolian transport is similar, however the former is more important for pollution Pb transport where the latter is more important for natural Pb. The fate of pollutant Pb is determined by future climate change. Wetter conditions could lead to a prolonged export of Pb further down-stream. If drier conditions prevail, eolian transport will be more important and pollution Pb could stay in soils.

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1. Introduction

Lead (Pb) occurs naturally in the environment as a lithogenic element, and its storages (pools) and pathways (fluxes) in a landscape vary with landscape development and environmental changes (in e.g. vegetation, eolian activity and precipitation) that affect erosion and weathering (Thevenon et al., 2010; Koinig et al., 2003). It is also regarded as a pollutant, being emitted from, e.g., mining, industry, and transport activities (leaded gasoline) causing deposition both locally (Thevenon et al., 2010; Bindler & Rydberg, 2016) and far from the emission source through long-range atmospheric transport (Bindler et al., 2001). Once deposited pollution Pb will be bound in organic soils or dissolved in water, as it has a strong affinity for organic matter (Walraven et al., 2014). Geogenic Pb, on the other hand, is bound to mineral particles and will not be soluble in water. This means that eolian transport will be more important (Rydberg et al., 2016).

The emissions of Pb peaked at around 1970, with anthropogenic emissions in the mid-1980s exceeding natural emissions by a factor of 27 (Nriagu, 1991). Thereafter Pb-emissions decreased rapidly, with a 93% reduction within EEA-33 countries (the countries that are members of the European Environmental Agency) between 1990 and 2017 (European Environmental Agency (EEA), 2019). The success is largely accredited to the large-scale legislative phaseout of leaded gasoline, in e.g. the Clean Air Act and its amendments in the US (Environmental Protection Agency (EPA), 1990) and the Directive on the Quality of Petrol and Diesel Fuels in the EU (Directive of the European Parliament and of the Council, 1998). Even though emissions of Pb has dropped, concentrations in sediment and soils remain elevated (Koinig et al., 2003; Thevenon et al., 2010; Bindler et al., 2001; Datko-Williams,Wilkie & Richmond-Bryant.,2013). Indeed, a study in a boreal catchment system shows that pollution Pb has a long residence time in soils (200-800 years from soil to water) indicating a prolonged period of impact from Pb pollution (Klaminder et al., 2006). The elevated concentrations and long residence time of Pb in soils suggests that many soils have acted as Pb-sinks. In some Scandinavian Arctic and Subarctic lakes, however, anthropogenic Pb-input to the aquatic system has remained elevated even though deposition has drastically decreased (Klaminder et al., 2010; Liu, Jiang, Zhang & Xu, 2011). This indicates that the Pb is derived from the catchment soils and hence, that some soils now act as Pb-sources.

There has been a considerable deposition of pollution Pb – emitted in industrial mid-latitudes – in the Arctic (Bindler et al., 2001). From the long residence-time of Pb in soils (Klaminder et al., 2006) along with this high deposition, it may be deduced that Arctic systems stores a sizable proportion of pollution Pb. Furthermore, the ongoing climate change and its effects on terrestrial and aquatic systems are predicted to be especially pronounced in Arctic regions (Kattsov et al., 2005; Vaughan et al., 2013). The temperature and melting season lengths are expected to increase, simultaneously as precipitation and evapotranspiration patterns are expected to change (Macdonald, Harner & Fyfe, 2005), with many Arctic areas likely becoming drier (Koenigk et al., 2013; Smol & Douglas, 2007). This may lead to changes in trace element storage and cycling (i.e. hydrological patterns and thawing of permafrost), as has been seen to be true for both Pb and mercury (Hg) at high latitudes (Klaminder et al., 2010; Rydberg et al., 2010). Thus, the understanding of Pb-pools and pathways in different Arctic systems is of interest to interpret patterns and predict future implications of climate change.

In this study, a catchment-wide mass balance model for Pb will be calculated for the Two-Boat Lake (TBL) catchment, i.e., a well-studied West Greenlandic lake catchment (in other literature also called SS903 or Talik Lake). The catchment is the focus of a long term (10 years) study of hydrology and biogeochemistry that has resulted in e.g. extensive

datasets and models (Lindborg et al.,2016; Johansson et al., 2015; Petrone et al., 2016), as well as a carbon mass-balance budget (Lindborg et al., 2020). The latter of which this study shares a similar approach. The purpose of the Pb mass-balance budget is to increase understanding of the transport and storage of Pb in a dry periglacial landscape which in turn enables discussions and implications of future changes (decreasing deposition and climate change). In this dry landscape, eolian transport is of great importance (Rydberg et al., 2016) and hydrological transport should be small when compared to wetter landscapes (Lindborg et al., 2020). Through the established mass balance model of TBL, the following questions will be answered; 1: How do pools and fluxes relate to each other and what does this mean? 2: Does TBL catchment soils act as a sink for atmospheric deposition or as a source to the aquatic system? 3: Is eolian or hydrological transport the most important pathway of Pb to the lake? and 4: How does the fluxes and pools of Pb in TBL catchment compare to those of a mid-latitude catchment? 5: How may the answer to these questions develop according to future climate change?

2. Methods

2.1. Study Site

The TBL catchment is situated in West Greenland, approximately 25 km from the settlement of Kangerlussuaq and 500 m from the Greenland ice sheet (at lat 67.125940, long −50.180370) (Figure 1). It is situated just above the polar circle and the area is characterized as having a dry polar tundra climate with local average annual precipitation and temperature of 269 mm (2011-2018) and -4.4 degrees Celsius (2011-2015), respectively (Johansson et al., 2015; Cappelen 2016). The area is underlain by a 300-400 thick continuous layer of permafrost, with permafrost-free through taliks forming under larger lakes (Harper et al., 2011). Despite being situated close to the ice sheet, and hence, being defined as proglacial, the lake lacks any hydrological connection to the ice sheet which means the lake is precipitation-driven and can also be defined as periglacial (Slaymaker, 2011).

The area of the catchment is 1.56 km^2 wherein the aquatic part (i.e. the lake) covers an area of 0.37 km^2. (Johansson et al., 2015). The soils of the terrestrial catchment are dominated by a layer of eolian silt with a mean thickness of 1 m (Petrone et al., 2016). In some low-lying areas where the conditions are wetter, the eolian deposits can be classified as peaty silt with a higher percentage of organic material (Rydberg et al., 2016; Petrone et al., 2016; Johansson et al., 2015). In some areas prone to erosion, the underlying soil which is dominated by till and glaciofluvial deposits is exposed (Petrone et al., 2016). The vegetation can be divided into dry grasslands, dwarf shrubs (Betula- or Salix-dominated), meadows, wetlands, and barren areas, where there is a void of vegetation (Lindborg et al., 2016; Clarhäll, 2011). The vegetation partly determines the thickness of the active layer, which ranges from 0.48 m in wetland and 0.81 m in grasslands (Table 1) (Petrone et al., 2016).

Table 1. Areas and active layer depth of vegetation types in the TBL catchment area.

Vegetation class	**Area (m²)**[1]	**Active layer mean depth (m)**[2]
Wetland	83 975	0.48
Dwarf shrub	162 334	0.7
Dry Grassland	231 655	0.81
Meadow	445 814	0.68
Barren	257 431	2[3]

[1] Values from Lindborg et al. (2016). [2] Values from Petrone et al. (2016). [3] Value from Harper et al. (2011).

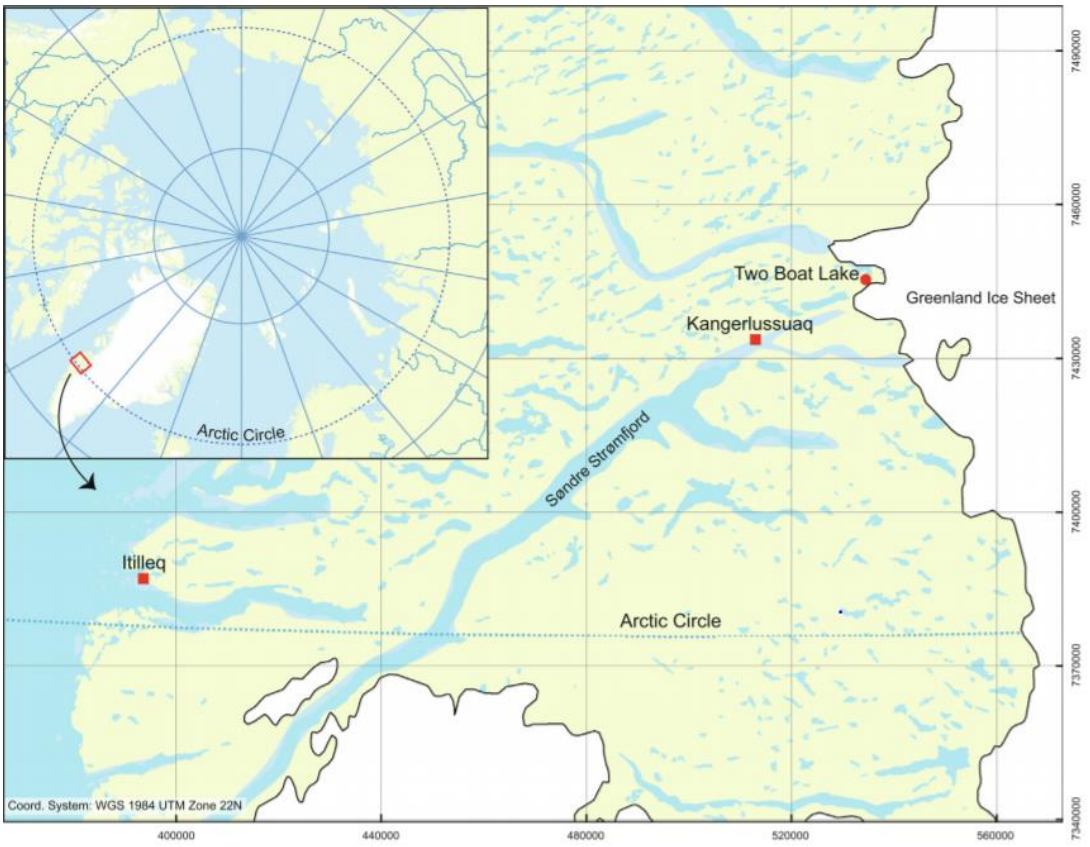

Figure 1. Map showing the location of TBL in West Greenland. Modification for Lindborg et al. (2016) after Johansson et al. (2015a).

2.2. Project Design

The design of this study shares the basis and reasoning of the carbon mass balance performed by Lindborg et al. (2020). The landscape is divided into different pools (e.g., lake water column, lake sediment, active layer soils, permafrost soils etc.) and fluxes (e.g., precipitation, runoff, sedimentation etc.) in a coupled terrestrial-aquatic conceptual mass balance model for the entire catchment (Figure 2). Pb-concentrations in soil, water and sediments (Lindborg et al., 2016) together with information on geometries (Petrone et al., 2016; Johansson et al 2015) as well as hydrological flows (Johansson et al., 2015), form the basis of the data used to calculate the mass balance. A summary of the sampling which was carried out from 2010 to 2018 and analyses performed by these authors are found in the following section 2.2. Calculations for the mass balance are described in section 2.5.

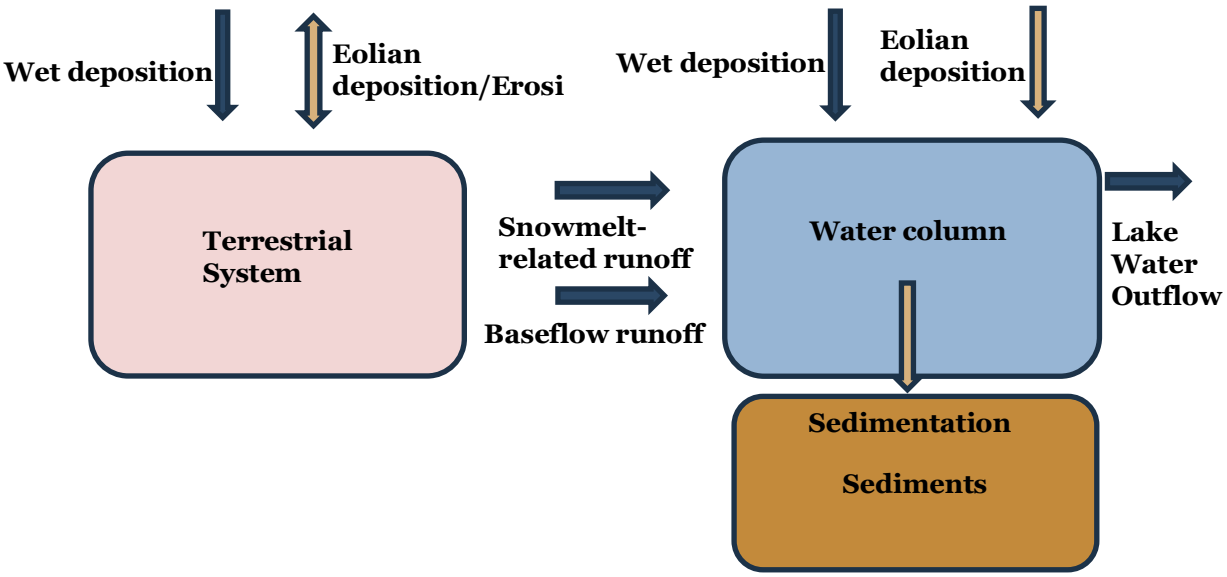

Figure 2. Conceptualized model of flows and major systems in TBL catchment.

2.3. Sampling

2.3.1. Atmospheric Deposition

Rain samples were collected and filtered in the summer during 2013 and 2014 with plastic pans (Lindborg et al., 2016). In April 2011 fresh snow were sampled along three transects using a standardized snow tube sampler (Lindborg et al., 2016). To acquire an estimation for organic eolian deposition, old snow in sheltered locations where eolian dust were clearly visible were collected in April 2012 and 2013.

2.3.2. Terrestrial System

In 2014 and 2015 short (0-12 cm) soil cores from each vegetation type (2-7 per type) were collected (Lindborg et al., 2016). From these, bulk density was determined for the different depths (Lindborg et al., 2016). Soil samples used for determining Pb-concentrations were collected in the eolian surficial layer (Lindborg et al., 2016). In August 2011 soil samples were collected with a steel cylinder with 10 cm intervals, from 10 cm below the surface to the permafrost. Additionally, in the same year, soil samples from different soils were taken with a steel spade (Lindborg et al., 2016). In 2018, a profile was sampled at a wetland site which extended down into the permafrost.

Terrestrial biota samples, from which Pb-concentrations are derived, were collected in august 2011 and 2014. Vegetation were sampled in different locales and muskox (*Ovibatus Moschati*) muscle tissue was made available through a local hunter (Lindborg et al., 2016).

2.3.3. Pore- and Surface Water

Porewater samples were collected using lysimeters installed in wetland areas during 2011-2018. Surface water samples from temporary streams, ice wedges and ponds were collected in 2012, 2014, 2017 and 2018 by hand-filling with a plastic flask. Both filtered (0.45 µm) and unfiltered samples were collected.

2.3.4. Aquatic system

Lake water samples were sampled with a Ruttner sampler from 0.5 m, 3 m, 5 m, and 25 m depths in various parts of the lake. Aquatic biota samples were collected in late summer

during 2011, 2012 and 2014 (Lindborg et al., 2016). The samples included aquatic vascular plants, algae, macrofauna and zooplankton collected from both the shore and deeper areas (using an Ekman grabber).

2.3.5. Sediment

13 surface-sediment cores (23-40 cm long) were collected in August 2012 (Lindborg et al., 2016). The locations of the cores were selected to represent different locations in the lake, different bottom types (vegetated or non-vegetated) and different water depths. The cores were sectioned into 1-cm sections for 0-10 cm depth, 2-cm sections for 10-20 cm and 4-cm sections for a core depth below 20 cm. Sediment accumulation rates of each core was estimated using C-14 and Pb-210 dating in macrofossils and bulk sediments respectively (Lindborg et al. 2016).

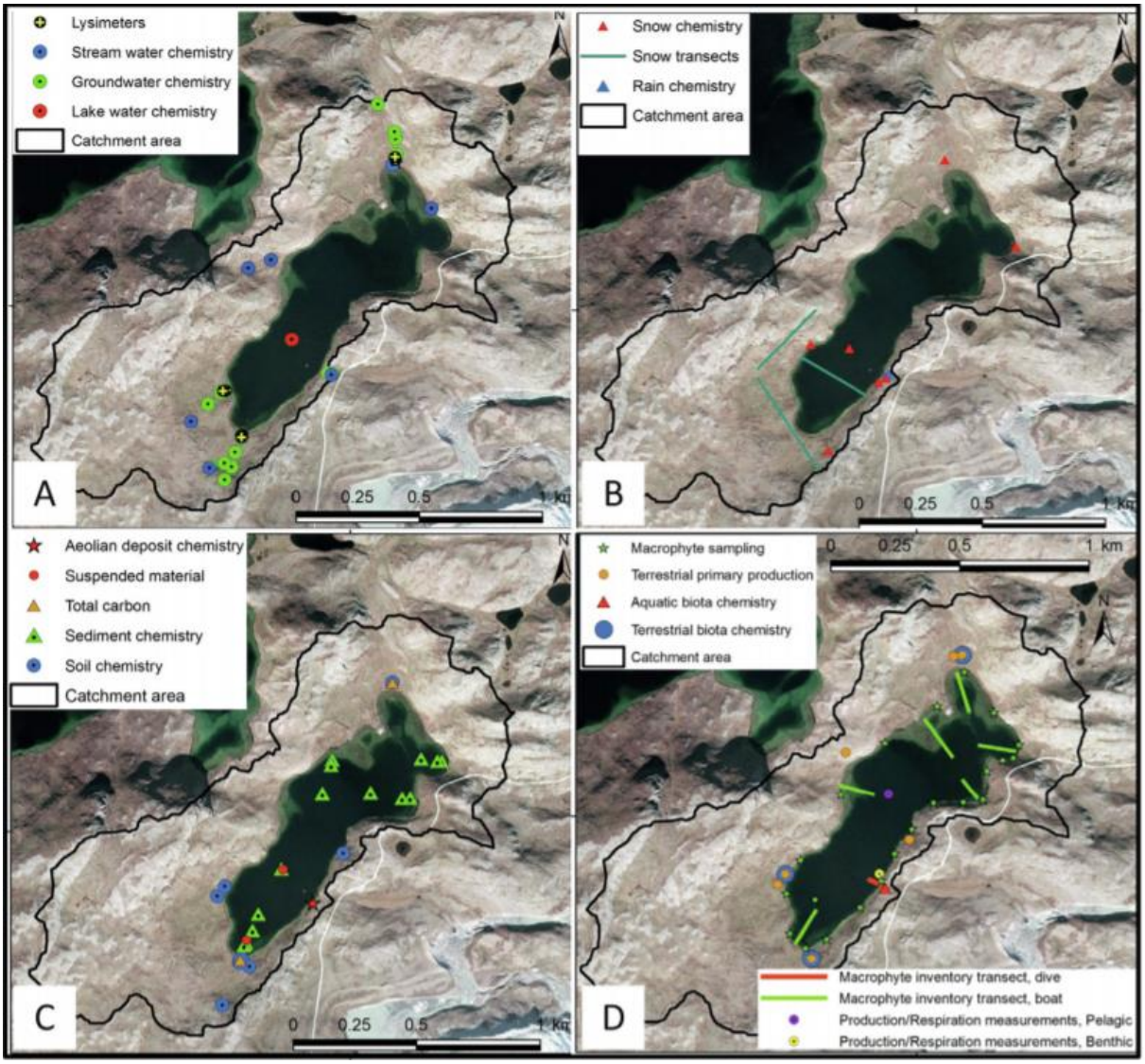

Figure 3. The TBL-catchment and sampling points for different factors. Modified version in Lindborg et al. (2020) from Lindborg et al. (2016).

2.4. Analyses

Pb-concentrations in liquid samples were determined using Inductively Coupled Plasma, Sector-Field Mass-Spectroscopy (ICP-SFMS; Lindborg et al., 2016). Pb-concentrations in solid samples were either analyzed using ICP-SFMS following digestion (Lindborg et al., 2016) or through wavelength dispersive X-ray fluorescence spectroscopy (WD-XRF) on loose-powder samples, following the method described by Rydberg (2014). Sediment

samples and the soil core extending into the permafrost were analyzed through WD-XRF while remaining solid samples were analyzed with ICP-SFMS. ICP-SFMS analyses were carried out at an accredited laboratory at ALS Scandinavia in Luleå. WD-XRF analyses were performed at the Department of Ecology and Environmental Science (EMG) at Umeå university.

2.5. Mass Balance Calculations

2.5.1. Atmospheric Deposition

Mean annual precipitation volumes (Johansson et al., 2015) and mean Pb-concentration in precipitation (Lindborg et al., 2016) were used to calculate annual Pb-flux for the terrestrial and aquatic part, respectively. The organic part of eolian deposition onto the lake was estimated using the old snow samples (corrected after sublimation (Johansson et al., 2015)) and calculated according to the following method described by Lindborg et al. (2020). The reasoning of this method is that eolian transport is higher during the winter and the area where snow accumulation occurs is estimated to be 20% of the total lake area (Johansson et al., 2015). Thus, 20% of the winter precipitation onto the lake was multiplied with the old snow Pb-concentrations. To account for eolian deposition made during the summer months, the winter eolian Pb-deposition was multiplied by a factor of 1.5 (Lindborg et al., 2020).

In estimating the minerogenic part of the eolian deposition, another approach was used. Zirconium (Zr), which was also analyzed in sediment and soil samples (Lindborg et al., 2016), does not weather to any noticeable extent (Middelburg, van der Weijden & Woittiez, 1988). Due to the lack of streams in TBL catchment, it can be argued that very little or negligible amounts of particulate Zr is transported with running water. This means that the only major input of Zr to the aquatic system should be that through eolian transport of mineral soils. Because of the particulate properties of Zr, all Zr that the lake receives should be incorporated into the sediment without major residence time in lake water and biota. According to this logic all Zr incorporated into the sediment each year derives from minerogenic eolian deposition, which enables Pb-input through the minerogenic part of the eolian deposition to be estimated using the Zr:Pb ratio in the eolian deposits (i.e. the source). Annual total Zr-input was calculated by first using the mean sediment accumulation rate of the 13 sediment cores (section 2.3.5) and the mean Zr-concentration of sediments. This value divided with the Zr:Pb ratio equals the estimation of annual Pb-input to the lake through minerogenic eolian deposition.

In Lindborg et al. (2020) it was argued that carbon input through eolian deposition onto the terrestrial system was negligible, because visible organic material would then accumulate which cannot extensively be seen. It can be argued that the same holds true for minerogenic elements such as Pb. Thus, eolian deposition and erosion is assumed to equal each other out.

2.5.2. Terrestrial Pool

The total Pb-pool in the terrestrial part of the catchment was divided into an active layer-, a permafrost-, and a terrestrial biota pool. Active layer depths for different vegetation types were determined by Petrone et al. (2016) (table 1) and these were divided into an upper (0-12 cm) and a lower section (12 cm-permafrost). This distinction was made due to high organic content in the topmost layer, and thus, a lower bulk density. Together with the areal extent of each vegetation type (Lindborg et al.2016) (table 1) the volume of the upper- and lower active layers for each type were calculated. Bulk densities (from Lindborg et al. (2016) for the uppermost layer were represented by the mean value of the measured cores (0-12 cm) while the bulk densities of the deeper layers were estimated by the deepest measured bulk density (at 12 cm). Bulk densities and volumes allowed for the total mass to be calculated for the two layers of each vegetation type. By multiplying these

masses with the Pb-concentrations of corresponding depths and vegetation type, Pb-pools for both sections of the active layer were calculated. By adding up the pools both layers of each vegetation type, the entire active layer Pb-pool was estimated.

The permafrost Pb-pool in the eolian layer was determined for each vegetation type in a similar fashion. The permafrost thickness (soil thickness (1m) – active layer thickness) were first calculated for each vegetation type. The thickness multiplied with the area of each vegetation type gave the eolian permafrost volumes of corresponding type. The deepest measured bulk density (at 12 cm) of the vegetation types were used to calculate the corresponding eolian permafrost total masses. Pb-pools of each vegetation type were calculated with the mass multiplied with corresponding mean Pb-concentrations of samples below the active layer. Lastly, the entire eolian permafrost Pb-pool were derived by adding up the pools of each vegetation type. lastly Pb-concentration values for samples below the AL of each vegetation type. For non-vegetated barren areas, values for dry grassland has been used in the calculations above (Lindborg et al., 2020). Pb in soils below the eolian layer is ignored here for two reasons. The first is that this layer is currently not near being incorporated with the active layer. Secondly it can also be argued that most Pb, and all pollution Pb, should be found in the upper layer. This partly due to pollution and partly due to the eolian supply of fresh mineral particles containing Pb.

The terrestrial Pb-pool in biota was calculated using already determined values of total carbon pools (both above and below ground) (Lindborg et al., 2020). For the vegetation pool, total carbon values of each vegetation type were converted into dry weight using the van Bemmelen conversion factor of 0.58 (van Bemmelen, 1890). The Pb-pool in terrestrial vegetation was then estimated using the total dry weight and the mean Pb-concentration of the analyzed terrestrial vegetation samples. The Pb-pool in herbivores were estimated using the same method, with total carbon values derived from Lindborg et al. (2020). Herbivore dry weight multiplied with Pb-concentration in muskox tissue gives the estimation of the Pb-pool in herbivores. Because of the very low Pb-concentrations in herbivores (Lindborg et al., 2016) removal of Pb through grazing was assumed to be negligible (almost all Pb is resupplied through feces).

2.5.3. Pore- and surface water pool and annual runoff

Soil water as well as annual runoff volumes (snowmelt-related surface runoff and baseflow runoff) (mean values from 2011-2015 including both wet and dry years) are presented in Johansson et al. (2015). T-test shows that porewater (associated with baseflow runoff) Pb-concentrations are significantly higher than stream water concentrations (associated with snowmelt-related surface runoff) (average values of 1.45 $\mu g\ l^{-1}$ and 0.22 $\mu g\ l^{-1}$ respectively, $p<0.05$). Thus, the flow of each type was calculated separately with their respective Pb-concentrations.

When calculating the annual runoff certain observations were taken into consideration. Namely that most of the runoff to the lake occurs during the snowmelt and that during this time there is an additional area (0.51 km^2), otherwise not hydrologically connected with the catchment, that contributes with runoff to the lake (Lindborg et al., 2020). Therefore, annual runoff calculations were divided into snowmelt-related surface runoff (55200 $m^{3)}$ and runoff during the rest of the year (baseflow runoff) (38600 m^3), as was done in Lindborg et al. (2020). The volumes of these are derived from Johansson et al. (2015) and the volume during the snowmelt were corrected for the extended catchment area in Lindborg et al. (2020). Total Pb transport into the lake were calculated with these volumes together with the mean Pb-concentrations of stream water (for snow melt runoff since streams only exist during this period and soil water generally is still frozen) and porewater (for remaining runoff).

2.5.4. Aquatic Pool and outflow

Maximum lake volume (4250000 m^3) (Johansson et al., 2015) together with mean lake water Pb-concentration (Lindborg et al., 2016) were used to calculate the non-biotic Pb pool in the water column. Lake water outflow only occurs periodically, but the mean annual volume is described in Johansson et al. (2015ab). This volume together with the lake water Pb-concentration was used to calculate annual removal of Pb from the aquatic system through the lake outlet.

The aquatic biota pool was calculated from already determined total carbon pools for various biota groups (Lindborg et al., 2020). Using different conversion factors, namely 0.4 for macrophytes (Kautsky, 1995), 0.3 for macrofauna (Kautsky, 1995), 0.5 for bacteria (Kemp et al., 1993), 0.48 for zooplankton (Lindborg et al., 2020) and 0.14 for phytoplanktion (mean value for different taxa from Olrik et al. (1998)), the total dry weight was calculated. The mean Pb-concentration value of all aquatic biota samples (Lindborg et al., 2016) together with the calculated dry weight was then used to estimate the total aquatic biota Pb-pool.

2.5.5. Sediment Pool and Accumulation

Studies has shown that the sediment accumulation rates in the lake has been spatially and temporally variable to a high degree (Rydberg et al., 2016; Bindler et al., 2001). A minimum and maximum value for sediment accumulation rate (Kg m^{-2} yr-1) were determined in Lindborg et al., (2020). These values, along with the area where sedimentation occurs (total lake bottom area – barren bottom area) and the mean Pb-concentration of lake sediment (Lindborg et al., 2016; Petrone et al., 2016) were used to calculate minimal and maximal annual Pb-removal from the aquatic system through sedimentation.

The whole basin mean sediment accumulation rate was determined using the average of the estimated rate from each of the 13 sediment cores (section 2.4.1) and the mean Pb-concentrations of sediments (Lindborg et al., 2016). The time during which sediment accumulation has occurred has been estimated to be roughly 4000 years (Rydberg et al., 2016). This allows for a rough estimation of total lake sediment Pb-pool to be made.

3. Results

There is in total roughly 14 500 kg (or ca 14.5 tons) Pb in TBL catchment (table 2). The majority of the Pb can be found in the terrestrial system, with approximately 13 500 kg Pb in total (94% of total Pb-weight) and a specific pool of 11 500 kg Pb km^{-2}. The terrestrial soils (active layer soils + permafrost soils) accounts for roughly 13 500 kg, which means that the total Pb-pool in biota and soil water is about 4 kg (<0.1% of total Pb weight). The Pb-pool of terrestrial soils is mainly constituted of active layer Pb, with roughly 10 700 kg (74% of total) as opposed to 2 800 kg (20% of total) in the permafrost. The aquatic system accounts for the remaining 910 kg Pb (6% of total) with a specific pool of 2 400 kg Pb km^{-2}. Less than 1 kg of Pb can be found in aquatic biota and lake water, hence almost all Pb in the aquatic system is stored in the lake sediments (Table 2).

On a whole-catchment scale, 111 g Pb (44 g Pb from precipitation and 67 g Pb from eolian deposition) enters the catchment annually while 150 g Pb (mostly through sedimentation) is removed (table 3, figure 4). This gives the entire system an annual balance of -39 g Pb. For the terrestrial system, input is the 34 g Pb yr^{-1} derived from precipitation and export to the lake is the 68 g Pb yr^{-1} leaving through hydrological runoff. The snowmelt-related runoff almost 5 times lower than that of the base flow, even though runoff volume during snowmelt exceeds that of the rest. Export is thus double the size of the input to the terrestrial system, which gives the terrestrial system a

negative balance with a deficit of -34 g Pb yr^{-1}. For the aquatic system, the inputs include the export from the terrestrial system (68 g Pb yr^{-1}) as well as 10 g Pb yr^{-1} through precipitation and 67 g Pb yr^{-1} is through eolian deposition (of which 60 g Pb yr^{-1} is minerogenic while 7 g Pb yr^{-1} is organic). Together, this gives an annual input to the lake of 145 g Pb. Removal of Pb from the aquatic system mainly consists of long-term storage through sedimentation (Low 39 g Pb yr^{-1}, High 510 g Pb yr^{-1} and mean 150 g Pb yr^{-1}), with only very limited removal through lake water outflow (0.15 g Pb yr^{-1}). This indicates that the aquatic system is in relative balance as 145 g Pb enters each year while 150 g Pb is removed (Table 3, figure 4).

Table 2. Calculated Pb-pools in different compartments of TBL catchment.

Pb-Pool	**Weight (kg)**	**Percent of total (aquatic+terrestrial) (%)**	**Area (km^2)**	**Specific Pools (Kg km^{-2})**
Terrestrial total pool	13 541	94	1.18	11 475
Active layer	10 691	74	1.18	9060
Permafrost (eolian layer)	2846	20	1.18	2410
Herbivore	0.0000067	<0.1	1.18	5.7E-06
Terrestrial plant biota	3.4	<0.1	1.18	3
Soil water	0.50	<0.1	1.18	0.4
Aquatic total pool	912	6	0.38	2400
Lake water	0.14	<0.1	0.38	0.4
Lake sediment	912	6	0.38	2400
Aquatic biota	0.19	<0.1	038	0.5
Total Pb-pool	14 453	100	1.56	11 500

Table 3. Calculated Pb-fluxes to and from the different compartments and the entirety of TBL catchment.

Flux	**Terrestrial System**	**Aquatic system**	**Entire System (Terrestrial+Aquatic)**
Area (m^2)	1 180 000	380 000	1 560 000
Precipitation ($g\ yr^{-1}$)	34	10	44
Precipitation ($\mu g\ m^{-2}\ yr^{-1}$)	29	26	28
Eolian organogenic deposition ($g\ yr^{-1}$)	0	7	7
Eolian organogenic deposition ($\mu g\ m^{-2}\ yr^{-1}$)	0	18	4.5
Eolian minerogenic deposition ($g\ yr^{-1}$)	0	60	60
Eolian minerogenic deposition ($\mu g\ m^{-2}\ yr^{-1}$)	0	158	38.5
Snow melt related runoff ($g\ yr^{-1}$)	-12	12	0
Snow melt related runoff ($\mu g\ m^{-2}\ yr^{-1}$)	-10	31	0
Baseflow runoff ($g\ yr^{-1}$)	-56	56	0
Baseflow runoff ($\mu g\ m^{-2}\ yr^{-1}$)	-47	147	0
Lake water outflow ($g\ yr^{-1}$)	-	-0.15	-0.15
Lake water outflow ($\mu g\ m^{-2}\ yr^{-1}$)	-	-0.4	-0.01
Mean annual sediment accumulation ($g\ yr^{-1}$)	-	-150	-150
Mean annual sediment accumulation ($\mu g\ m^{-2}\ yr^{-1}$)	-	-395	-96
Total in ($g\ yr^{-1}$)	34	145	111
Total in ($\mu g\ m^{-2}\ yr^{-1}$)	29	380	71
Total out ($g\ yr^{-1}$)	-68	-150.15	-150.15
Total out ($\mu g\ m^{-2}\ yr^{-1}$)	-57	-395.4	-96.01
Annual balance ($g\ yr^{-1}$)	-34	-3	-35.15
Annual balance ($\mu g\ m^{-2}\ yr^{-1}$)	-28	-15	-25.01

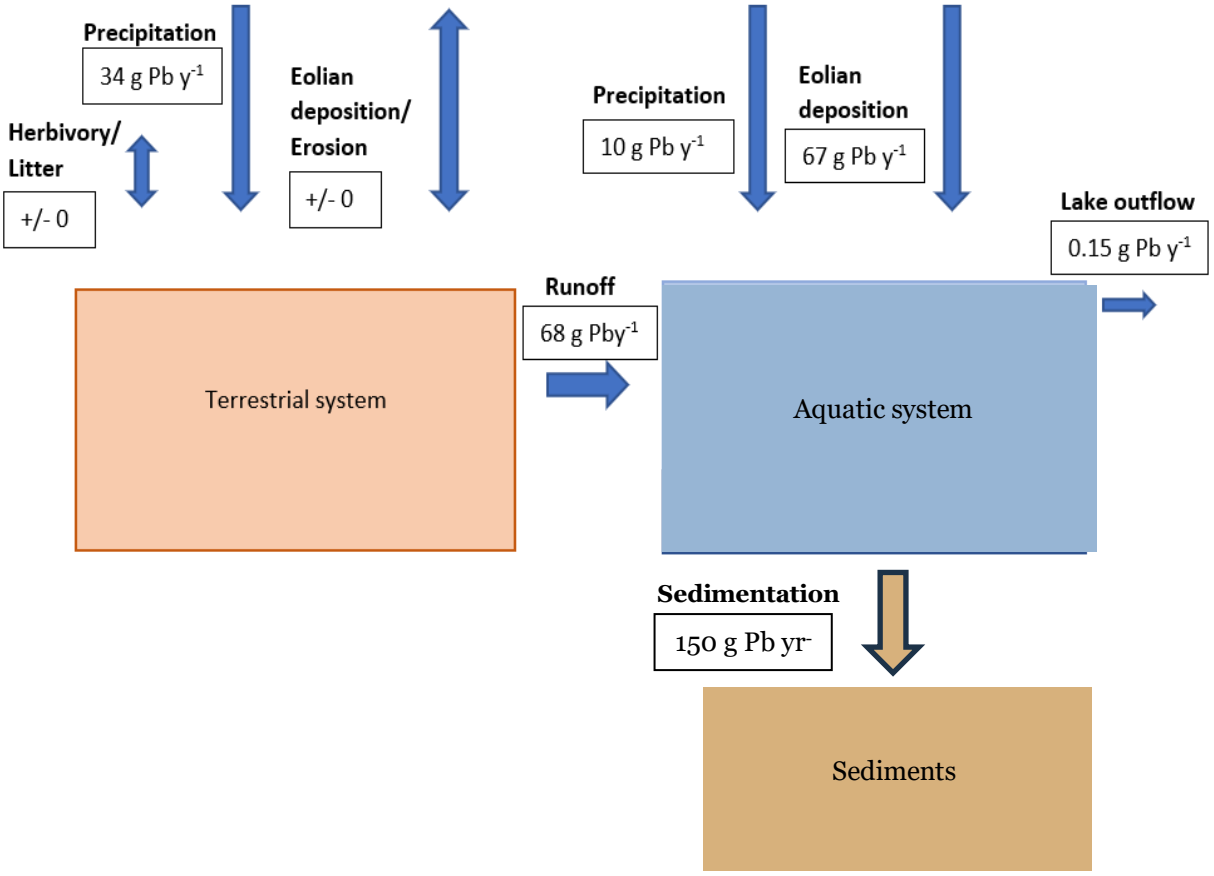

Figure 4. Conceptual figure of calculated fluxes in the TBL catchment.

4. Discussion

4.1. Pools and fluxes

That 94 % of Pb is stored in catchment soils (table 2) agrees with the studies that show that the residence time of Pb in soils are long and that levels remain heightened after decreased deposition (Koinig et al., 2003; Thevenon et al., 2010; Bindler et al., 2001; Datko-Williams,Wilkie & Richmond-Bryant.,2013; Klaminder et al., 2006). The fact that the majority of soil-Pb is stored in the active layer (10 691 kg of 13 537 kg), is probably partly explained by the active layer being larger but also by Pb-concentrations being greater (Lindborg et al., 2016). This makes sense because the active layer is where pollution Pb would be stored and hence would give the active layer a Pb-surplus. In the calculations of Pb-pools in soils, there are some uncertainties. For example, an eolian soil layer of 1 m is assumed universally for the catchment. Furthermore, the soil samples with Pb-concentrations of different vegetation types are in some cases few, which means that some accuracy might be missed. The Pb-pool in soils should hence be viewed as an approximation.

The Pb-pool in biota is considerably lower when compared to that of soils (<0.1% of total catchment Pb as compared to 94%). This is explained by the much lower volume (especially pronounced due the dry tundra environment in TBL) and lower Pb-concentrations in biota (Lindborg et al., 2016). The same reasoning may explain the low Pb-levels in soil water (porewater), as the volume is small compared to soils (especially

pronounced due to the dry conditions of TBL) and that only dissolved organic Pb constitutes the Pb-pool, meaning minerogenic Pb is missing. Uncertainties in calculated biota values include: few samples being used (for herbivores), not having a full coverage of plant species and the usage of a conversion factor (van Bemmelen, 1890). These Pb-pools, however, are so small that improved accuracy would not alter the result in a noteworthy way. Fluxes to and from the terrestrial system are small when compared to the total pool (e.g. annual wet deposition only accounts for 0.0003% of total Pb-pool in soils) (table 3). This may also be because of these flows being hydrological, which means that mostly the organic fraction of Pb is transported in this way (due to the lack of streams with particulate transport). The much higher concentrations of Pb in porewater as compared to stream water (Lindborg et al., 2016) is most likely explained by the former being in contact with soils (with a large pool of Pb) for an extended time , while latter is directly derived from precipitation. Hydrological sampling and modeling (Johansson et al., 2015) are relatively extensive and well-constrained, which means that uncertainties in pools and fluxes in water probably are more accurate. Some uncertainties, however, might originate from temporal variation being missed due the sampling not being continuous (Johansson et al., 2015).

In the aquatic system nearly all Pb is stored in the lake sediment, which accounts for 6% of entire catchment Pb (Table 2). As also discussed for the terrestrial system, this could be due to sediments incorporating both minerogenic and organic Pb (while biota and water mainly incorporates organic Pb). Furthermore, the sediment may be argued to be the final terminus of all Pb in TBL catchment. This due to terrestrial Pb eventually leaching to the aquatic system and the lack of outflow meaning that the majority of Pb will end up here. The high Pb-content in sediments is also reflected by the relatively high input of eolian minerogenic deposition (60 g Pb yr^{-1} of total 145 g Pb yr^{-1}) (table 3), due to this deposition being directly incorporated into the sediment (as discussed in section 2.5.1.). Organic eolian deposition, originating from e.g. erosion scars from within and outside the catchment, will probably to a higher degree be dissolved in lake water (Heindel et al., 2017). The majority of eolian input to the lake most likely originating in glacial outwash plains (i.e. being minerogenic) (Rydberg et al., 2016), explains the much higher eolian input of minerogenic Pb than organic (table 3). Additional uncertainties arise when calculating aquatic pools and fluxes. Among them is the dating that determines the sediment accumulation rate from which annual Pb-removal through sedimentation is calculated (Lindborg et al., 2016). The organic eolian deposition is also uncertain, due to the old snow sample not necessarily representing the deposition and due to high variability in eolian activity (Rydberg et al., 2016). Another major uncertainty lies in the aquatic biota pool. This is mostly because of the many different conversion factors used.

4.2. Mass-balance budget; Pb-sink or source?

The negative balance of the terrestrial system (-34 g Pb y^{-1}) means that simultaneously as deposition is of decreasing importance, a relatively large transport to the aquatic system still occurs. This means that even though the effects of decreased deposition can be seen in the terrestrial system the same cannot be said for the aquatic system, which is also the case in a Norwegian Arctic lake (Liu et al., 2011). Along with decreased deposition (Bindler et al., 2001) and subsequent slow leaching from the terrestrial system, the negative balance could also partly be explained by potential thawing of permafrost. The current episode of warming has led to a decrease in permafrost size in several Arctic locations (Lyon et al., 2009; Rydberg et al., 2010). This means Pb leached from the terrestrial system will become mobile and further add to the negative balance. However, Pb stored in permafrost will probably mostly be minerogenic (due to the active layer containing most pollution Pb) which means transport to the aquatic system would be limited.

The hydrological input to the lake is considerably larger than wet deposition input (68 g pb yr^{-1} as compared to 10 g Pb yr^{-1}) (table 3). This has also been seen in various U.K. lakes

(with lesser importance of eolian activity), where 97.6% of Pb entering lakes derive from hydrological transport from the catchment (Yang et al., 2018). Even though most eolian deposition input originates from outside the catchment (Rydberg et al., 2016), there should at least be some transport of Pb from the terrestrial system of TBL to the lake. This would mean that the soils of TBL catchment currently act as Pb-sources to the lake as opposed Pb-sinks, which has been previously suggested in earlier stages of pollution. Contemporary soils that are Pb-sinks are still found in some English heavily industrial areas (Shotbolt, Rothwell & Lawlor, 2008; Nicholson et al., 2003) indicating continued local pollution problems or low mobility in said soils. The mass-balance budget is also negative when viewing the whole catchment as one system (-39 g Pb yr^{-1}) (Table 3). However, the export of Pb from the catchment is almost only that through sedimentation. This means that the catchment in its whole currently acts as a Pb-sink, as removal in this way can be argued to be permanent (Lindborg et al., 2020).

4.3. Eolian vs hydrological transport

Eolian and hydrological transport to the lake is of almost equal magnitude, 67 g Pb yr^{-1} and 68 g Pb yr^{-1} respectively. This means that the pattern differs from that of carbon in the same catchment (Lindborg et al., 2020), where hydrological carbon input is much greater than eolian input. The reason for this difference might be the inclusion of minerogenic Pb-input in this study, a type of eolian deposition with low carbon levels (therefore excluded in Lindborg et al. (2020)). Lake sediment deposited during periods of high eolian activity has high Zr-values (Rydberg et al., 2016), which indicates a high minerogenic Pb input and thus validates its inclusion.

The fates of hydrologically transported Pb as compared to that of eolian Pb may differ. The majority of eolian input is minerogenic Pb bound with mineral particles. These will, as previously discussed (section 2.5.1.), directly be incorporated in the sediment. Dissolved Pb that is being transported hydrologically however, would have the ability to be transported further down-stream as well as being incorporated in biota. Hence, as of now, eolian and hydrological transport is of near equal importance (due to low lake outflow). If lake outflow were to increase however, this might change. Another difference between hydrological and eolian flows might be in its origins. Minerogenic eolian Pb from outwash plains (majority of eolian transport) is natural, whereas much of the dissolved Pb in water can be argued to derive from pollution (due to pollution Pb binding to organic matter (Walraven et al., 2014) and subsequent leaching). Thus, to follow the fate of pollutant Pb and its inclusion into biota, the hydrological flows and the climate changes´ impact on them is of great importance.

4.4. Comparison with mid-latitude catchment

When comparing the Pb mass-balance of TBL to that of the English Howden catchment (Shotbolt, Rothwell & Lawlor, 2008), some major differences can be seen. The catchment is larger than TBL (35.5 km^2 as compared to 1.56 km^2), is dominated by organic-rich soils and has riverine input into a reservoir within the catchment. Most prominent of the differences from TBL catchment are the hydrological flows, where e.g. wet deposition is calculated to roughly 3000 μg m^{-2} yr^{-1} in Howden catchment as compared to 28 μg m^{-2} yr^{-1} in TBL. Consequently, the hydrological export to the lake is 840 μg m^{-2} yr^{-1} compared to 59 μg m^{-2} yr^{-1}. This makes sense due to the dry conditions around TBL, its lack of riverine transport and due to the local pollution sources of Howden catchment (Shotbolt, Rothwell & Lawlor, 2008).

Even though Howden catchment terrestrial soils have a positive Pb balance (more Pb enters the soils than what is exported, i.e. the soils act as a sink) where TBL has a negative (soils act as a Pb-source), the latter has a higher specific pool of Pb in soils (roughly 13.5 tons Pb km^{-2} in TBL as compared to 8.64 tons Pb km^{-2} in Howden catchment). One reason for this is that only a 20-cm soil depth has been used for Howden catchment (Shotbolt,

Rothwell & Lawlor, 2008) as compared to TBL. However it is further explained by hydrological flows being of greater importance in Howden catchment, meaning great quantities of organic and even minerogenic (if streams are strong enough to transport mineral particles) Pb can be exported from the terrestrial system. Thus, a continuous export of Pb from terrestrial soils has been active that may have removed substantial parts of previously deposited Pb. The dry conditions of TBL catchment in contrast, means that nearly all minerogenic Pb and much of the organic Pb remains in soils (low hydrological transport) simultaneously as there is a major supply of fresh eolian minerogenic Pb from distant sources (not existing in Howden catchment).

4.5. Future development of system

With time, it is plausible that organic Pb (to some extent derived from pollution) stored in soils will slowly be transported into the lake sediment. Thus, if atmospheric deposition continues to decrease, future disposition of Pb in TBL catchment may be slightly more focused in lake sediments instead of terrestrial soils (as pollutant Pb now bound to organic matter in terrestrial soils will eventually leach into the aquatic system). Until this might occur, the negative balance of the terrestrial system will likely persist. Furthermore, if the current warming continues, the permafrost will continue to thaw and consequently the active layer size will increase. This means that the terrestrial system most probably will act as a source of pollutant Pb to the aquatic system for some time to come.

How climate change will affect the pathways of Pb in the catchment is somewhat uncertain. If precipitation increase it may be argued that runoff to the lake will increase and that this will be the most important Pb-input to the aquatic system. Increased runoff and precipitation could lead to an increase in lake water outflow, perhaps leading to TBL catchment in its whole becoming a Pb source. This would mean that Pb from TBL catchment would spread to down-stream systems and eventually the ocean. However, increasing temperatures will probably also lead to higher rates of evapotranspiration. This would instead imply that conditions will be unchanged or even drier. If drier conditions prevail, as is expected in many Arctic locations (Koenigk et al., 2013; Smol & Douglas, 2007), eolian transport would instead be enhanced due to higher mobility and availability of material (Lancaster, 2013). Eolian supply is already of major importance to the area's lake sediment composition (Rydberg et al., 2016) and will probably be even more pronounced in the future if the area becomes drier. Thus, in years to come, minerogenic eolian deposition might be the largest input of Pb to the lake. Some things that needs to be taken into consideration, however, is that organic eolian deposition will eventually run out of Pb (due to much of it being pollution Pb) and that minerogenic eolian supply might decrease due to increased distance to glacial outwash plains (retreating ice-front).

4.6. Uncertainties and inaccuracies

The Pb mass-balance budget for TBL catchment builds upon several assumptions and simplifications. This means that it does not reflect reality completely accurately but instead functions as a conceptualized model. Major uncertainties lie in the calculations of organogenic eolian deposition and annual sedimentation. The former is based on accumulated organic material in old snow that necessarily is not representative (Lindborg et al., 2020). Furthermore, eolian activity is not constant (Rydberg et al., 2016; Heindel et al., 2017) which means a longer period of sampling would be required for more accurate values. However, the organogenic part of eolian deposition, is according to the calculations of lesser importance. This means that the uncertainties should not affect the overall accuracy too much. Sedimentation calculations builds upon sediment cores where spatial and temporal variations are taken into consideration, thus increasing the accuracy. The dating, however, that serves as the foundation for sediment accumulation rate calculations introduce some uncertainties. This is partly due to the rough estimation of the depth that corresponds to 150 years age (Lindborg et al., 2020). Minor inaccuracies for i.e. hydrological values may also derive from temporal variations that might be missed due to

sampling-periods that are too short (Lindborg et al., 2020). Even though the values might not be entirely accurate; they are plausible when compared to other systems (section 4.3) and put into context. The mass-balance model should hence at least have some degree of conceptual value from a discussion standpoint.

4.7. Conclusion

Most Pb is stored in terrestrial soils (94%), while 6% of total catchment Pb is in lake sediments. Other Pb-pools are small in comparison (<1% combined). The TBL-catchment acts as a Pb-sink in its entirety, partly due to its limited outflow. The soils, on the other hand, are major sources. Eolian and hydrological transport to the lake are currently of equal magnitude, however eolian transport is suspected to be more important in the future if drier conditions prevail. This would mean that minerogenic eolian deposition would constitute the largest input of Pb to the lake. When compared to a mid-latitude catchment, Pb-fluxes in hydrological pathways are much less pronounced. This is believed to be attributed to the dry conditions of TBL catchment and the lack of local pollution sources. The dry conditions could also explain the high Pb-levels in soils, as Pb deposited over a long period of time is very slowly exported to the aquatic system. If climactic changes are ignored, a prolonged period of organic Pb-transport from the terrestrial system to the lake sediments is expected (so that much of previously deposited pollutant Pb will eventually be stored in lake sediments). A future scenario with wetter conditions would mean that the pollutant Pb could spread further down-stream. Even dryer conditions would lead to much of pollutant Pb remaining in soils.

The mass-balance model of Pb in TBL catchment is a conceptualized model containing inaccuracies that could be improved upon in future studies. It can, however, be used to get an overview of Pb occurrence in a system and allows for discussions on its properties and implications of future change. Thus, a similar approach might be utilized to examine other toxic metals, like cadmium (Cd) and zinc (Zn).

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