

Mercury (Hg) Transport in a High Arctic River in Northeast Greenland

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Abstract In a warming climate, mercury (Hg) pathways in the Arctic can be expected to be affected. The Hg transport from the high arctic Zackenberg River Basin was assessed in 2009 in order to describe and estimate the mercury transported from land to the marine environment. A total of 95 water samples were acquired and filtered (0.4 μm pore size), and Hg concentrations were determined in both the filtered water and in the sediment. A range of other elements were also measured in the water samples. Hg concentrations in the filtered water were in general highest in the beginning of the season when the water came mainly from melted snow. THg concentrations in the sediment were in general relatively constant or

slightly decreasing until mid-August, where after the concentrations increased. A principal component analysis separated the samples into spring, summer and autumn samples indicating seasonal characteristics of the patterns of element concentrations. The total amount of Hg in the sediment transported was estimated to 2.6 kg. Approximately 60% of the sediment-transported Hg occurred during a 24-h flood in the beginning of August caused by a glacial lake outburst flood. The total amount of transported dissolved Hg was estimated to 46 g, and 13% of this transport occurred during the 24-h flood. If it is assumed that the Hg transport by Zackenberg River is representative for the general glacial rivers in East Greenland, the total Hg transport into the North Atlantic from Greenland alone is approximately 4.6 tons year⁻¹ with an estimated annual freshwater discharge of $\sim 440 \text{ km}^3$.

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1 Introduction

Mercury (Hg) in the Arctic environment has long been an issue of great focus mainly because Hg constitutes a health threat for human populations exposed to Hg through their traditional diet (Johansen et al. 2004). Mercury is a naturally occurring element in the Arctic and is released through weathering of

rocks and as a result of volcanic/geothermal activities that constitute the primary natural sources (Munthe et al. *in press*). However, human activities have led to a considerable increase in Hg concentration in the Arctic environment compared to preindustrial times. Dietz et al. (2009) estimated that the anthropogenic contribution in wildlife hard tissue matrices was above 92%. Although the Hg emissions to the air has decreased in North America and Europe, it is still increasing in Asia (Munthe et al. 2011) and is likely to continue the increase in the coming decades (Streets et al. 2009).

The Arctic can act as a sink for the global Hg cycle (Schroeder et al. 1998; Lindberg et al. 2002; Lu et al. 2001), e.g. through Mercury depletion events (MDE). This phenomenon where gaseous elemental mercury (GEM) is transformed and removed from the atmosphere on the scale of hours or days during springtime was first reported by Schroeder et al. (1998). However, knowledge gaps around this theory have resulted in several debates on the importance and significance of MDEs versus net Hg loadings and other processes (Poissant et al. 2008). Most of the deposited Hg is re-emitted to the atmosphere (Munthe et al. 2011), but it is generally believed that a significant amount of Hg is deposited in the Arctic and subsequently methylated (Steffen et al. 2008).

Rivers also supply mercury to the Arctic marine coastal environment (Andersson et al. 2008; Graydon et al. 2009), but only a few studies estimating the amount of Hg transported from the terrestrial and freshwater systems to the Arctic coastal marine areas exist (Sukhenko et al. 1992; Coquery et al. 1995; Outridge et al. 2008). In a warming climate, this transport of Hg may change. Increasing or changing precipitation patterns may influence the Hg deposition from the atmosphere and the river water discharge. Similarly, increasing temperatures will likely influence freshwater and terrestrial processes known to be connected to the Hg pathway, e.g. methylation of inorganic Hg to methyl Hg (Macdonald et al. 2005), which is much more bio-available and hence will enter the food web. Melting of permafrost (Macdonald et al. 2005) and increased freeze-thaw events are processes that may also mobilise Hg to the environment. Therefore, it is uncertain how the Hg pathways and total transport in the Arctic will be affected (Macdonald et al. 2005).

The main aim of this study is to estimate the annual amount of total Hg (THg) that is transported from the terrestrial area of the Zackenberg River drainage basin in northeast Greenland to the marine environment. In the Zackenberg area, a comprehensive monitoring programme (ZERO; <http://www.zackenberg.dk>) has been running since 1995 with the purpose of describing the entire ecosystem and study how structure and function of the abiotic and the biotic environment respond to climate variability and change. The Hg results will be used in a comparison with existing published data from other Arctic rivers to improve the knowledge of Arctic annual land-to-sea transport.

2 Material and Methods

2.1 Study Site

Zackenberg River is located on Wollaston Forland in northeast Greenland at 74°N, 21°W (Fig. 1). The river drains a 483-km² basin where the western part is dominated by the A.P. Olsen Glacier and the St. Sødal Valley, which is surrounded by mountains based on gneiss and carbonate bedrock. The eastern part of the basin is almost without glacial influence, and the mountains around the Lindeman River valley and the Zackenberg Valley are based mainly on sandstone. The study area is high Arctic with a mean annual temperature of −9.0°C, and only June, July and August have average monthly temperatures above 0°C (Sigsgaard et al. 2010). Average annual precipitation is 261 mm, and more than 80% falls as snow during the 8–9 winter months (Hansen et al. 2008). The area has continuous permafrost and an active layer with thawing of the upper 45 to 80 cm every summer (Christiansen et al. 2008). Most vegetated areas are found in the Zackenberg Valley below 300 m a.s.l. and in the lowest lying areas of St. Sødal. The vegetation cover consists mainly of dry and moist dwarf shrub heath (e.g. *Dryas* sp., *Cassiope tetragona*, *Salix arctica*), wet grasslands, and fens (e.g. *Eriophorum triste*, *Duponthia psilosantha* and *Eriophorum scheuchzeri*) that covers around 40% of the valley area (Elberling et al. 2008).

The study was conducted during the summer 2009. The 2009 season was characterised by an unusual low amount of snow and a very early snow melt compared to previous years. The limited

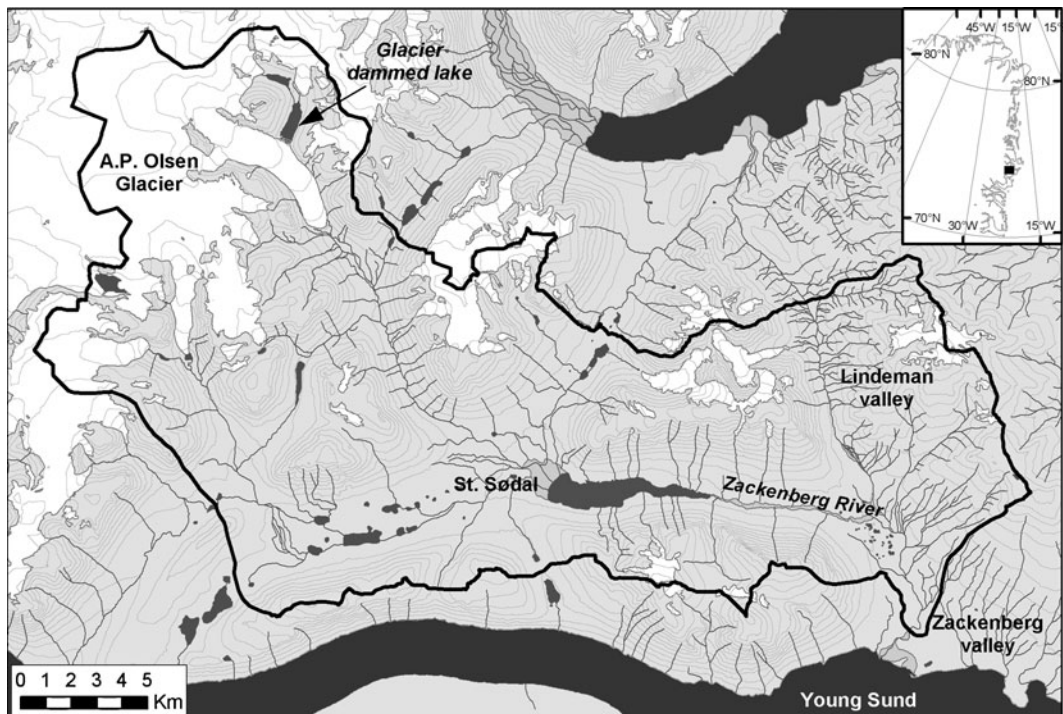


Fig. 1 The Zackenberg drainage basin and location of major valleys. The outlet of Zackenberg River into Young Sund is in the *lower right part of the area*

amount of snow combined with a relatively warm spring made snow disappear very early. The Zackenberg River broke up on 22 May, which is more than a week earlier than observed before. A major peak in water discharge was observed on 11–12 August when a flood caused by an outburst of a glacier-dammed lake raised the river water level by 1.5 m (Fig. 2). By the end of September, the river was again covered by ice, and only a limited base flow persisted. River depths were measured every 15 min from 22 May until 13 October using a SR50 Sonic Range sensor (Campbell Scientific). However, the Qh relation (relation between water level and discharge) needed to convert the water depth to discharge is only valid for a consistent river profile. In the spring when there was ice on the bottom of the river profile and snow on the banks as well as after the flood when the river profile changed, the previously established Qh relation could not be used. Discharge is therefore interpolated between manual discharge measurements during these periods.

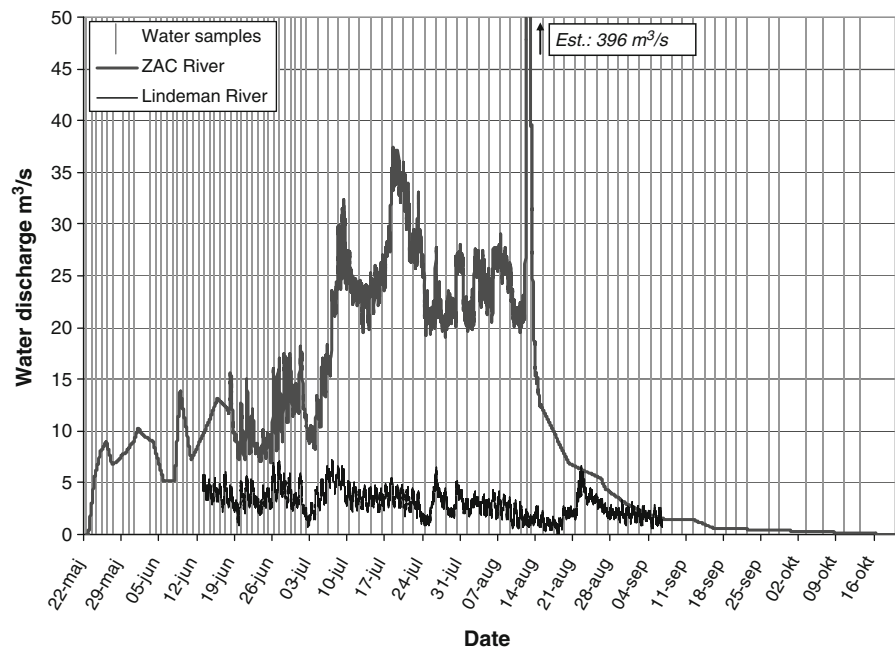
Lindeman River is a tributary to Zackenberg River and only drains a small part of the total drainage area. The discharge is therefore low without glacial lake

outburst floods (GLOFs) (Fig. 2). River depth in Lindeman is measured continuously with a Mini-Diver (Schlumberger Water Services) that has been corrected for barometric pressure and converted to discharge through six manual discharge measurements and a known Qh relation. The increase in discharge of the Lindeman River in late July and late August corresponds to rain events. This last rain event occurred after the flood when the Qh relation of the Zackenberg River is no longer valid, and it is therefore not visible in the Zackenberg River discharge patterns (Fig. 2).

2.2 Sampling

During the period from 22 May to 13 October 2009, a total of 95 depth-integrated water samples for Hg analysis were collected from the Zackenberg River, while the samples from Lindeman were collected when manual discharge measurements were performed. Sampling was performed daily at 8:00 AM. In addition, nine of the 95 samples were collected between 21:00 on 11 August and 20:00 on 12 August when the flood occurred. Approximately 250 ml

Fig. 2 Water discharge from Zackenberg River and the sub-discharge of the Lindeman River in 2009. Vertical lines mark time of water sampling for Hg analyses



water was filtered through Nuclepore 0.4- μm -pore-size filters into acid-cleaned glass bottles. The filters with sediment were dried and sent together with the filtered water samples to the laboratory at NERI, Denmark (sampling procedure is described in more detail in the GeoBasis manual; available at <http://www.zackenberg.dk/Monitoring/GeoBasis/>). Filters from samples of the first melt water did not have enough sediment for the analysis, and hence, only 85 sediment samples were analysed.

Furthermore, additional depth-integrated water samples, 248 samples in total, were collected by the GeoBasis monitoring programme at 8:00 and 20:00 every day, and the pH and conductivity were measured. The samples were filtered (Whatman GF-F; 0.7 μm pore size) and dried in order to determine suspended sediment concentrations (SSC).

2.3 Chemical Analyses

The 85 filtered sediments were analysed for THg using a Solid Sample Atomic Absorption Spectrometer, AMA-254 (Advanced Mercury Analyser-254 from LECO, Sweden), where Hg is led through several heating phases and finally measured by UV absorption.

The 95 water samples were analysed for THg following the US-EPA Method 1631, Revision E

(US-EPA 2002). The procedure is in short: To the water sample, a drop of BrCl solution is added to oxidise the Hg species to Hg^{2+} . The surplus of oxidants is removed by a drop of hydroxyl/ammonium, after which Hg^{2+} is reduced by a SnCl_2 solution and the released $\text{Hg}(\text{g})$ are driven by an Argon air current to a gold trap, where Hg is pre-concentrated. After collection for a pre-set time, the Hg is thermally desorbed into the atomic fluorescence detector (P.S. Analytical Millennium Merlin, Kent, UK). Detection limits, calculated as three times the standard deviations on blind samples, are typical 0.1–0.2 ng L^{-1} . The method is controlled by an independent quality control sample at 5 ng L^{-1} after standardisation and for every approximate 10 samples. Furthermore, the method is verified by participation in QUASIMEME laboratory intercalibration tests twice a year, where the results are usually within one standard deviation from the assigned mean values. The Hg concentration range tested in QUASIMEME is from 1.7 to 32 ng L^{-1} (Table 1).

In addition to Hg, a range of other elements were measured in the water samples using Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7500ce) at NERI, Denmark. The elements analysed include Li, Na, Mg, Al, P, S, K, Ca, Sc, T, V, Cr, Mn, Fe, Ni, Co, Cu, Zn, Rb, Sr, Y, Zr, Ag, Sn, Ba, La, Ce, Nd, Ta, Pt, Au, Pb, Bi and Th. The

Table 1 Results from NERI's participation in the QUASIMEME laboratory intercalibration study for Hg in seawater during the analysis period

Sample	Measured value	Assigned value±standard error	Z score	Analysis date
QTM148SW	3.57	4.40±0.65	-1.3	October 2009–January 2010
QTM149SW	6.07	6.98±0.97	-0.9	October 2009–January 2010
QTM150SW	31.09	32.37±4.15	-0.3	October 2009–January 2010
QTM154SW	1.70	1.92±0.34	-0.7	April–August 2010
QTM155SW	7.62	7.61±1.05	0.0	April–August 2010
QTM156SW	13.70	14.84±1.96	-0.6	April–August 2010

The Z scores of the results are the number of assigned standard errors between laboratories above or below the assigned value, i.e. Z score of less than two is within 95% confidence limit of all participating laboratories (10 to 16 results reported)

laboratory at NERI is accredited to ICP-MS analyses of the following elements in fresh water with the specified precision: V, Cr, Fe, Sr, Ba: 5%; Na, Mg, Al, K, Co, Ni, Cu, Cd, Pb: 10% and Li, P, Mn, Zn, Mo: 15%.

2.4 Data Treatment

The THg transport was calculated based on the concentrations of THg in both water and suspended sediment in the daily water samples multiplied with the daily water discharge. For days when THg concentrations were not measured, the mean THg concentration of the two nearest measurements (one before and one after) was used. During the 24-h flood on 11–12 August, the estimation procedure was based on measurements every 2–3 h instead of daily data. Although the pore size differed between the filtering of the water used for Hg analyses (0.4 µm) and the daily water samples used to estimate the suspended sediment concentrations in the river (0.7 µm), we assumed that these data could be combined without correction in order to estimate the amount of THg transported by the river. The amount of suspended sediment in the water is so high that the amounts of sediment on the two different sized filters in practice are the same. This was confirmed by filtering half of 30 samples taken at the same time in the Zackenberg River through the two different filters and weighing the amount of sediment on each filter. This test showed no significant difference between the two filter types (paired *t* test; *p*=0.59).

The patterns of element concentrations in the filtered water were analysed by a principal component analyses (PCA). The PCA was performed on standardised element concentrations, and 95% confidence

ellipses around seasons were drawn. The statistical analyses were performed using the statistical package R (R Development Core Team 2008)

3 Results

3.1 Seasonal Pattern of Discharge and Sediment Transport

Due to the sparse amount of melt water from snow in the Zackenberg Valley in 2009 and the low temperatures in the spring, the runoff in June was lower than ever recorded in the Zackenberg River since the environmental monitoring began in 1995. The total runoff in 2009 was also low compared to previous years and estimated to $144 \times 10^6 \text{ m}^3$ during the period 22 May–20 October. In the beginning of the season, water came mainly from melted snow, while later in the season, rain events contributed significantly to the water discharge of the Zackenberg River. Two major peaks in suspended sediment concentrations (SSC) were observed during the season. The first peak (3.053 mg L^{-1}) was measured on 4 August in the morning. This was not related to high water flow but probably originated from sudden landslides/erosion or block slumping along the river banks. The other peak in SSC (3.965 mg L^{-1}) was measured on 11 August (23:00) during peak flow of the 24-h glacial lake outburst flood (Sigsgaard et al. 2010).

3.2 Seasonal Pattern of THg Concentrations

The THg concentrations in filtered water and in suspended sediment in the Zackenberg River through

the 2009 season are shown in Fig. 3. In both the water and in the sediment, THg concentrations showed high day to day variations. THg concentrations in the filtered water were in general highest in the beginning of the season and decreased towards the mid-season. Concentrations decreased from approximately 0.6 ng L^{-1} at the end of May to 0.3 ng L^{-1} at the later part of June. Hereafter, concentrations stayed at approximately the same level until mid-October, although large fluctuations between days were measured. THg concentrations in the sediment were in general relatively constant or weakly decreasing until mid-August, where after the concentrations increased. From mid-August, temperatures at higher elevations fell below freezing, and hence, melting from the A.P. Olsen Glacier at the end of St. Sødal declined. Relative more water in the Zackenberg River therefore came from the Lindeman River than from St. Sødal at the end of the season (Fig. 2). Since Lindeman River drains an area with sandstone bedrock in contrast to gneiss and carbonate bedrock

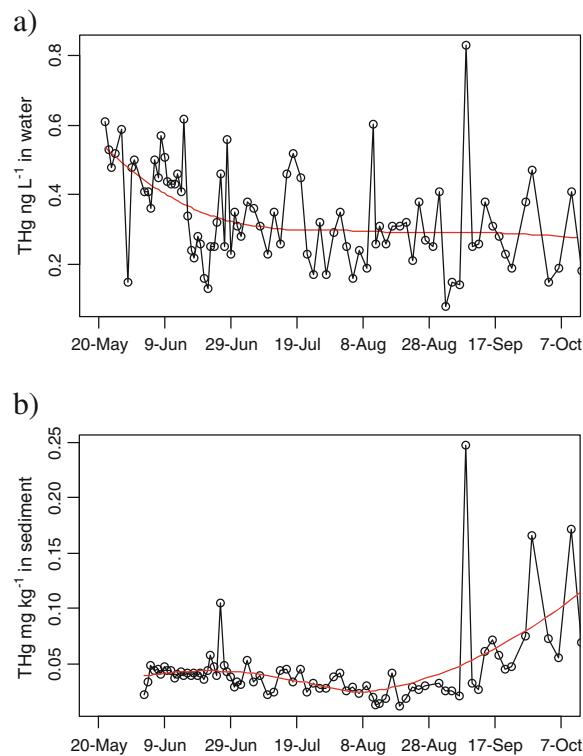


Fig. 3 THg concentrations in filtered water (ng L^{-1}) (a) and in retained suspended sediment (mg L^{-1}) (b) through the 2009 season in Zackenberg River. The red line represents a loess (local polynomial regression fitting) smoother line

in St. Sødal, this may have caused the changes in THg concentrations during the later part of the season.

3.3 Seasonal Pattern of Selected Element Concentrations

In addition to Hg, a range of other elements were analysed in the filtered water, and a PCA including the following elements Hg, Li, Na, Mg, Al, S, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Sr, Y, Zr, Ba, La, Ce, Nd, Pb and Th was performed. Because the levels of the elements were different by orders of magnitude, the concentrations were log-transformed, and the PCA was performed on the correlation matrix. The two first principal components explained 37% and 27% of the total variation. The PCA figure (Fig. 4) is a so-called biplot showing loadings of the elements at the first two components. The elements Al, Nd, Y, La, Pb and Ce had relative high positive loadings in Component 1 and low loadings in Component 2. Hg plotted close to these elements in the PCA figure. The elements were characterised by relative high concentrations in late May and the first weeks of June when the main snow cover was still melting (Fig. 2). Snow melt after this date originates mainly from the last deep snow patches. The element Sc was in contrast to this group and showed the opposite seasonal pattern with lowest concentrations until mid-June and higher in the rest of the season. Elements such as Ca, Na, S and Sr had relative high positive loadings at Component 2 and low loadings at Component 1. The seasonal pattern of these elements was characterised by a peak just before and after the 24-h flood in the beginning of August and with generally higher concentrations in the second half of the season compared to the first half of the season. The other elements were spread as a continuum between these two main groups of elements. The pH of the Zackenberg River was almost constant throughout the season ($\text{pH } 6.6 \pm 0.1$). Based on the elements relative concentrations the PCA separated the water samples into seasons, especially the spring samples were separated from the summer and autumn samples (Fig. 5).

3.4 The 24-H Flood and the Total THg Budget for the Zackenberg River

On 11 August, a flood event lasting approximately 24 h occurred in the Zackenberg River. The flood was

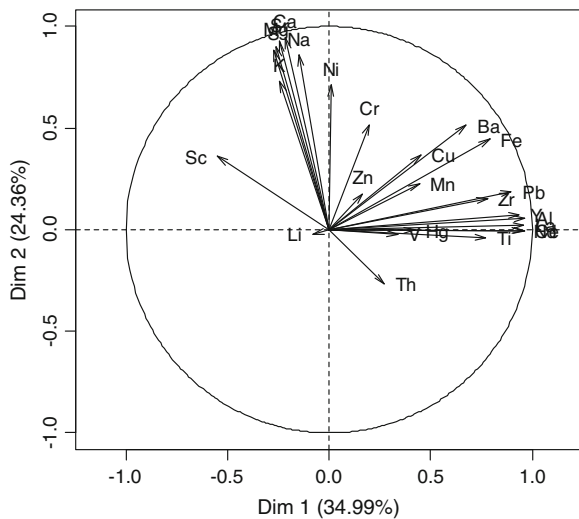


Fig. 4 Principal component biplot showing elements loadings

caused by an emptying of a glacier-dammed lake at A.P. Olsen Glacier (Fig. 1). The water discharge during the glacial lake outburst flood was less than 5% (approximately $7 \times 10^6 \text{ m}^3$) of the total annual discharge during the season but around 30% of the total suspended sediment transport (approximately 20,000 tons) occurred during this flood event. THg concentrations in filtered water during the 24-h study showed no clear patterns compared to the days before, and after, except that in the beginning of the flood, relative high concentrations were observed. THg concentrations in the sediment showed in general lower levels during the flood than in the days before

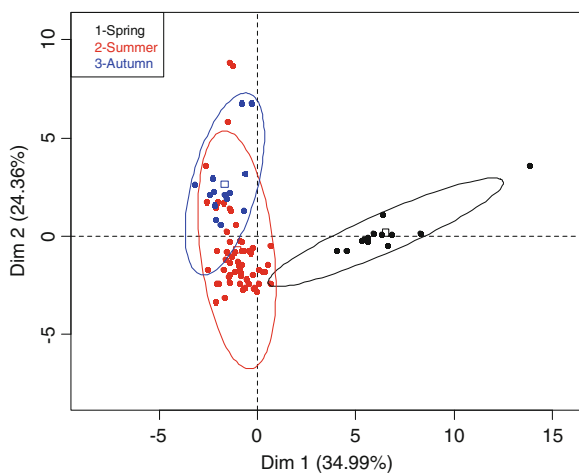


Fig. 5 Principal component 1 versus principal component 2 for the individual water samples. *Ellipses* represent 95% confidence limits for each season. *Squares* show ellipse centres

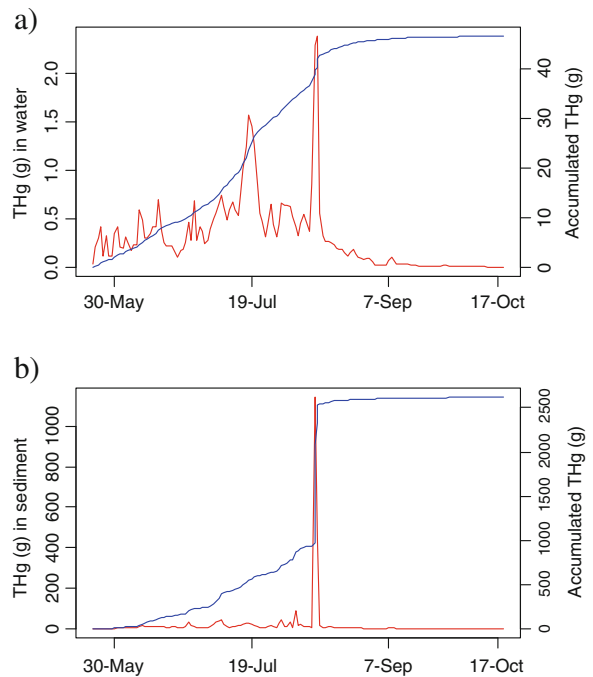


Fig. 6 The daily (*red*) and accumulated (*blue*) amount of THg transported by Zackenberg River as dissolved in the water (**a**) and in the suspended sediments (**b**) throughout the 2009 season

and after. The total amount of Hg in the sediment transported by the Zackenberg River was estimated to 2.6 kg (Fig. 6). Approximately 60% of the Hg transported in sediment occurred during the 24-h flood in the beginning of August. The total amount of dissolved Hg (Hg in water filtered to $<0.4 \mu\text{m}$ in size) was estimated to 46 g and 13% of this occurred during the 24-h flood.

4 Discussion

The dissolved THg concentrations in the Zackenberg River were slightly elevated in the beginning of the season. This may result from atmospheric Hg deposition during the winter and/or from the process of the atmospheric MDE. MDE has been reported to occur throughout the Arctic, sub-Arctic and Antarctic coasts (Steffen et al. 2008), and it is likely that MDE also occurs in the Zackenberg area and thereby contribute to the observed THg in the melted snow water. However, a high correlation between Hg and rare earth elements (REEs) such as La, Nd and Ce, which also showed the highest concentrations in the

beginning of the season, suggests that other processes may also be responsible for the elevated dissolved Hg concentrations during the early season. A similar peak in REEs during spring time in areas affected by freezing temperatures has previously been reported as a result of flushing of the upper thawed soil layer (Shiller 2010), which indicate that the dissolved THg could also come from this source.

About 50 times higher amounts of THg were transported in the suspended sediment phase of the highly glacier-influenced Zackenberg River than as THg dissolved in the water. This shows that the far most important factor for the transport of THg from the terrestrial and freshwater environment to the coastal areas is sediment transport. With the expected warming of Greenland and Arctic as a whole (ACIA 2005; IPCC 2007), more freshwater will flow from the glaciers through the terrestrial areas to the marine environment. This will likely lead to higher transport of sediments originating directly from enhanced melting of glaciers but also indirectly from erosion of river banks during more frequent high discharge events like the glacial lake outburst flood observed in this study. Furthermore, both precipitation and temperature in the Zackenberg area are expected to increase significantly in the future as a result of global warming and enhanced as the sea ice diminishes (Stendel et al. 2008), and this will likely further

increase the transport of terrestrial THg to the marine environments.

Estimated Hg concentrations from five Arctic rivers and their transport of Hg are shown in Table 2. The data foundation for these estimations are very inhomogeneous, spanning from one sample event in 1 year as was the case for the three large Russian rivers (Lena, Ob and Yeisei Rivers) to comprehensive sampling throughout the seasons over 3 years in the Mackenzie River. The dissolved Hg concentrations in the Zackenberg River were a factor of 10 lower, while Hg concentrations in the sediment were comparable with those reported. The total THg transport was considerably lower from the Zackenberg River compared to the five other rivers due to a much lower annual water discharge.

Sigsgaard et al. (2010) report the annual discharge and suspended sediment transport for the Zackenberg River for 1997–2009. The average annual discharge for this period is 35% higher than for the year 2009 (194 mill m³ compared to 144 mill m³). However, the average annual suspended sediment transport for the 1997–2009 period is only slightly lower (7%) than for 2009. The reason for this is believed to be based in the occurrences of the GLOF. In years where the GLOF occurred during summer, where the surface layer and sediments on the river banks had melted, the sediment transport was considerably higher than in

Table 2 Comparisons of Hg transport by different Arctic rivers

River	Water discharge (km ³ year ⁻¹)	Sediment discharge (10 ⁶ tons year ⁻¹)	Mean Hg concentrations		Hg transport			Reference
			Hg _D (ng L ⁻¹)	Hg _P (μg g ⁻¹)	Hg _D (kg year ⁻¹)	Hg _P (kg year ⁻¹)	Hg _T (kg year ⁻¹)	
Zackenberg	0.144	0.052	0.34±0.15	0.04±0.03	0.046	2.6	2.646	This study
Lena	525	17.6	5.0±0.6	0.12±0.12	1,150	2,900	4,000	Coquery et al. (1995)
Ob	429	16.5	2.8±0.6	0.05±0.003	530	820	1,300	Coquery et al. (1995)
Yeisei	620	5.9	1.5±0.7	0.05±0.007	410	310	700	Coquery et al. (1995)
Katun, West Siberia	17.5	2.6	0.8	1.35	1,300	3,500	4,800	Sukhenko et al. (1992) ^a
Mackenzie river	330	124	1.61	0.013	532	1,637	2,169	Outridge et al. (2008) ^b

Hg_D dissolved Hg (Hg in water filtered to <0.4 μm), Hg_P particle-bound Hg (>0.4 μm), Hg_T=Hg_D+Hg_P

^a Mean of 2 years

^b Based on the original study by Leitch et al. (2007)

years with no GLOF or only GLOF during the frozen period (e.g. November 2008).

Merrild et al. (2008) estimated an average annual freshwater discharge of $\sim 440 \text{ km}^3$ from East Greenland to the Greenland–Iceland–Norwegian Seas during the period 1999–2004. This estimation was based on a snow model combined with in situ data from two long-term meteorological and hydrometric stations. If it is assumed that the Hg transport by the Zackenberg River is representative for the total discharge from East Greenland, the total Hg transport can be estimated to approximately $4.6 \text{ tons year}^{-1}$ using the suspended sediment transport in the Zackenberg River for the corresponding years (1999–2004) and the estimated 2009 Hg transport. Although this up-scaling is a rough estimate not taking into account the difference in Hg transport from various sources of freshwater, e.g. calving, glacial subflow, runoff from non-glaciated areas etc., we consider this as a first estimation, which has to be improved by future research. Outridge et al. (2008) advanced a mass balance for Hg in the Arctic Ocean. Based on data from the three large Russian rivers and the Mackenzie River (Table 2) together with an estimate of the total water discharge to the Arctic Ocean, they estimated a total riverine Hg flux of $\sim 12.5 \text{ tons year}^{-1}$. The abovementioned rivers accounted for approximately $8.3 \text{ tons year}^{-1}$ and adding our estimate for the East Greenland area of $\sim 4.6 \text{ tons year}^{-1}$ exceed the estimate of total riverine Hg flux to the Arctic Ocean by Outridge et al. (2008), which indicate that the $12.5 \text{ tons year}^{-1}$ may be an underestimate.

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