



# Distribution of rare earth elements and other high field strength elements in glacial meltwaters and sediments from the western Greenland Ice Sheet: Evidence for different sources of particles and nanoparticles

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## ABSTRACT

Although global warming increases meltwater input into the oceans, rather little is known about the distribution of high field strength elements such as the rare earths and Y (REY), Zr, Hf, Th, and U in arctic glacial meltwaters and glacial-fed rivers. We studied glacial meltwater and glacial-fed rivers from the Kangerlussuaq area and a glacial-fed lake and its meltwater inflow in the Isua area, both in the western part of the Greenland Ice Sheet (GRIS). Trace element concentrations were determined in 0.2  $\mu\text{m}$ -filtered water samples (“dissolved” fraction), in the respective filter residues (“particulate” fraction) and in ambient sediments (including cryoconite from a meltwater pond on the surface of the GRIS). We also measured “truly dissolved” REY concentrations in a 10 kDa-ultrafiltered sample from glacial-fed Watson River.

Shale-normalized (“ $\text{SN}$ ”) REY patterns of the particulate fraction and ambient sediments (including cryoconite) are rather similar to those of local Archean gneisses and show positive  $\text{Eu}_{\text{SN}}$  anomalies. This rules out Asian dust (which does not show positive  $\text{Eu}_{\text{SN}}$  anomalies) as a possible source of these aluminosilicate particles, but suggests that local Archean basement is eroded and currently transported by ice, water and wind to the depositional sites in front of and onto the GRIS. All 0.2  $\mu\text{m}$ -filtered glacial-fed rivers show very unusual  $\text{REY}_{\text{SN}}$  patterns in comparison to tropical, temperate and boreal rivers, and are significantly enriched in light relative to heavy REY. For glacial-fed Watson River, the <10 kDa-ultrafiltrate shows much lower REY concentrations than the <0.2  $\mu\text{m}$ -filtrate, suggesting that >99% of La and >78% of Yb in the latter are associated with nanoparticles and colloids. Although the  $\text{REY}_{\text{SN}}$  patterns of the <0.2  $\mu\text{m}$ -filtrates are rather similar to those of the particulate fraction and ambient sediments, they lack any  $\text{Eu}_{\text{SN}}$  anomalies, whereas the <0.2  $\mu\text{m}$ -filtrates from a meltwater pond on the surface of the GRIS show positive  $\text{Eu}_{\text{SN}}$  anomalies similar to (but somewhat smaller than) the cryoconite. While the water from the meltwater pond carries nanoparticles derived from local Archean sources, the lack of  $\text{Eu}_{\text{SN}}$  anomalies in the glacial-fed rivers suggests that their nanoparticle and colloid load represents atmospheric dust that is remobilized from greater depth in the GRIS. Most of this dust probably originated from eastern Asia and shows a REY distribution similar to Post-Archean upper continental crust, i.e., lacks any  $\text{Eu}_{\text{SN}}$  anomaly. Hence, REY geochemistry suggests that the particulates and the nanoparticles/colloids in these arctic meltwaters are derived from different sources.

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## 1. Introduction

Global warming accelerates the retreat of the polar ice caps and of glaciers (Barry, 2006; Dyurgerov, 2002; Dyurgerov and Meier, 1997a,b) and enhances input of glacial meltwaters into the oceans. The Greenland Ice Sheet (GRIS) suffers from significant ice mass loss (IPCC, 2007; Ramilien et al., 2006; Velicogna and Wahr, 2005) that is not presently compensated by increased precipitation. The seasonal runoff of the GRIS is heavily influenced by variations in meteorological

conditions and its melting effect (Mernild and Hasholt, 2009). For instance, the maximum discharge shows high variations between 2007 and 2010, with  $1430 \text{ m}^3 \cdot \text{s}^{-1}$  in 2007,  $550 \text{ m}^3 \cdot \text{s}^{-1}$  in 2008,  $680 \text{ m}^3 \cdot \text{s}^{-1}$  in 2009 and  $1620 \text{ m}^3 \cdot \text{s}^{-1}$  in 2010 for the Watson River at Kangerlussuaq (Hasholt et al., 2012). On top of seasonal variations there exists a general trend of increasing ice mass loss of the GRIS since 1990, primarily caused by increased runoff and melting (Ettema et al., 2009; Tedesco et al., 2013). Despite the increasing importance of glacial meltwater input into the oceans and in marked contrast to input via tropical, temperate and boreal rivers, rather little is known about the concentrations, distribution and behavior of trace elements in arctic glacial meltwaters and glacial-fed rivers. However, as the

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trend of significantly increasing runoff from the GRIS into the ocean is very unlikely to change anytime soon, it is important to better characterize the trace element geochemistry of glacial-fed rivers which drain the GRIS.

There is still controversy concerning the origin of dust in and on the GRIS. While several studies (e.g., Biscaye et al., 1997; Svensson et al., 2000) suggested that most of the dust present in ice cores originated from Asia (e.g., from the Chinese Loess Plateau), other studies (e.g., Wientjes et al., 2011) have presented evidence that at least the dust component in cryoconite from the dark region of the western ablation zone is locally derived material with admixed anthropogenic components.

The rare earths and Y (REY) show coherent geochemical behavior in natural systems due to their very similar physico-chemical properties. While in natural systems most REY are exclusively trivalent, Ce and Eu are redox-sensitive and may be tetra- and divalent, respectively, and then behave differently from the strictly trivalent REY. Systematic variations within the trivalent REY result from the decrease of their ionic radii with increasing atomic number, and minor fractionation between neighboring isoivalent REY may occur due to subtle differences of the stabilities of some of their chemical complexes.

Until now, the only available data for the rare earth element (REE) distribution of lakes and rivers in Greenland were for a single sample from Lake Isua in western Greenland (Goldstein and Jacobsen, 1988a), which is a terminal glacial meltwater lake. Lake Isua shows a shale-normalized (sub-script SN; shale is Post-Archean Australian Shale, PAAS, of McLennan, 1989) REE pattern with enrichment of the light REE (LREE) relative to heavy REE (HREE). Compared to other pristine rivers and lakes worldwide this is a highly unusual REE distribution (Fig. 1). However, the arctic Great Whale River from Quebec, Canada, also shows a LREE-enriched pattern (Goldstein and Jacobsen, 1988a, b). Hence, these two data sets may indicate that the dissolved REE inventory of arctic glacial-fed rivers may differ significantly from that of other river waters.

Here we discuss the distribution and behavior of trace elements, in particular of the REY and other particle-reactive trace elements such as Zr, Hf, Th, and U, in glacial-fed rivers, in a meltwater stream on the

surface of the GRIS, in a terminal lake, and in a freshwater lake in western Greenland. We report trace element data for samples taken in August 2013 in the Kangerlussuaq area in western Greenland and in 2010 from Lake Isua and from the glacial outflow into Lake Isua, some 150 km north of Nuuk. We discuss the concentrations and distributions of these elements in 0.2  $\mu\text{m}$ -filtrates (the “dissolved pool”), in the respective filter residues (the “particulate pool”), in associated sediment, and in one ultrafiltered (10 kDa) sample (the “truly dissolved pool”) from the mouth of the Watson River in Kangerlussuaq. We use the REY as proxies to trace the origin of the sediments, particulates and nanoparticles/colloids and show that nanoparticles/colloids in the glacial-fed rivers are derived from “old” Asian dust remobilized from deeper levels in the GRIS, whereas the cryoconite, the river sediments and the river particulates reflect the Archean bedrock that is currently eroded and deposited by waters and winds on and around the GRIS.

## 2. Study area and methods

### 2.1. Study area and samples

Our samples originate from the area between Kangerlussuaq (67°00' 30.96"N; 50°41'21.38"W), Russell Glacier (67°05'29.61"N; 50°14' 16.38"W) and parts of Israndsdalen (67°06'54.92"N; 50°10'22.79"W) and from a sampling location on the GRIS (67°09'54.92"N; 50°10' 22.79"W). Other sampling sites are in the Isua region (65°11'12.30"N; 49°51'53.44"W) further south, some 150 km north of Nuuk (Figs. 2; S1).

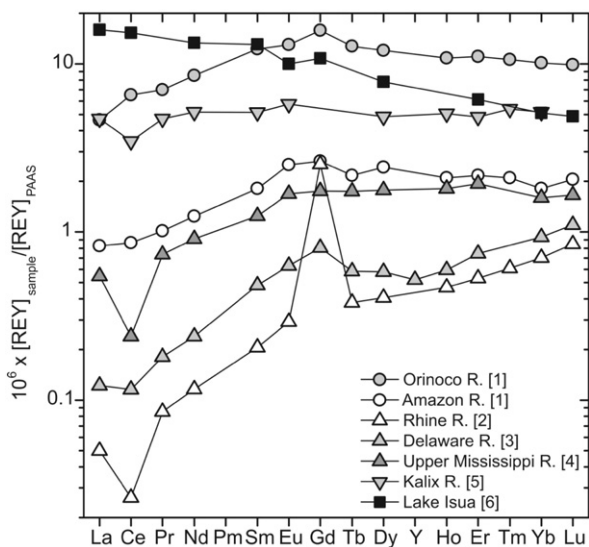
River water samples were taken in August 2013 from the Qinnguata Kuusua River (unnamed glacial catchment approximately 7 km south of Russell Glacier), at several sites from the Akuliarusiarsuup Kuua River (at Israndsdalen, at Russell Glacier, and ca. 1100 m upstream from the confluence of the Qinnguata Kuusua and Akuliarusiarsuup Kuua rivers; Russell Glacier catchment) and from the Watson River in Kangerlussuaq, ca. 2500 m downstream from the confluence of the Akuliarusiarsuup Kuua and the Qinnguata Kuusua rivers and some 500 m before the river enters the Søndre Strømfjord and mixes with seawater. For comparison, lake water without any direct input of glacial meltwater was sampled at Lake Ferguson (Tasersuatsiaq; 66°58'55.16"N, 50°41'35.91"W). Two meltwater samples were taken from below a thin ice cover on a shallow meltwater pond on the GRIS at 67°09'54.92"N and 50°10' 22.79"W. For comparison to published data, two samples from the Isua region in West-Greenland were sampled during an unrelated field campaign in July 2010. One water sample was directly taken from the outflow of the glacier (65°11'29.24"N, 49°51'49.93"W) into Lake Isua, and another one was taken from terminal Lake Isua proper (65°11'12.30"N, 49°51'53.44"W).

For convenience, we will refer to samples from the Qinnguata Kuusua River, the Akuliarusiarsuup Kuua River, the Watson River, Lake Isua and the glacial outflow into Lake Isua as “glacial-fed”, to the sample from Lake Ferguson, as “lake water”, and to the meltwater samples from the surface of the GRIS as “pure meltwater”. The term “glacial-fed” is commonly used to characterize rivers in Iceland (Louvart et al., 2008) and will also be applied here to rivers in Greenland.

Whenever possible, sediment samples were collected from the river bed or the river bank. Sediment from the shallow meltwater pond was scraped from the bottom of the pond.

### 2.2. Analytical methods

Water samples were taken and stored in acid-cleaned 1000 mL polyethylene bottles and were kept cold and dark until further treatment. The samples were filtered through 0.2  $\mu\text{m}$  cellulose acetate membranes (Sartorius). The residual particles on these filter membranes will be referred to as “filter residues”. The filtrates were then acidified to pH 1.8–2.0 with 6 M HCl before 0.5 ml of a 100 ppb Tm spike was added as an internal standard. An aliquot of the sample solution was separated and used to determine trace element concentrations by Inductively



**Fig. 1.** REY<sub>SN</sub> patterns of tropical, temperate, boreal and arctic rivers. Data for Orinoco River and Amazon River from Deberdt et al. (2002) [1], Rhine River from Kulaksiz and Bau (2011) [2], Delaware River from Bau et al. (2006) [3], Upper Mississippi River from Shiller (2002) [4], Kalix River from Ingri et al. (2000) [5] and glacial-fed Lake Isua from Goldstein and Jacobsen (1988a) [6]. Note that the positive Gd anomaly for the Rhine River is anthropogenic and that more recent data also shows anthropogenic La and Sm anomalies (Kulaksiz and Bau, 2013).

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