



# Stable isotopic evidence for anaerobic maintained sulphate discharge in a polythermal glacier



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## ABSTRACT

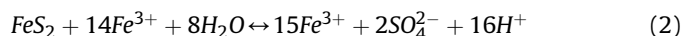
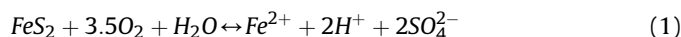
To understand the sources and sinks of sulphate and associated biogeochemical processes in a High Arctic environment, late winter snowpacks, the summer melt-waters and rock samples were collected and analysed for major ions and stable isotope tracers ( $\delta^{18}\text{O}$ ,  $\delta^{34}\text{S}$ ). The  $\text{SO}_4^{2-}/\text{Cl}$  ratio reveal that more than 87% of sulphate (frequently > 95%) of total sulphate carried by the subglacial runoff and proglacial streams was derived from non-snowpack sources. The proximity of non-snowpack sulphate  $\delta^{34}\text{S}$  ( $-8$ – $-19\text{‰}$ ) to the  $\delta^{34}\text{S}$  of the major rocks in the vicinity ( $-6$  to  $+18\text{‰}$ ) suggest that the non-snowpack sulphate was principally derived from rock weathering. Furthermore,  $\text{Ca}^{2+} + \text{Mg}^{2+} / \text{SO}_4^{2-}$  molar shows that sulphate acquisition in the meltwaters was controlled by two major processes: 1) coupled-sulphide carbonate weathering (molar ratio  $\sim 2$ ) and, 2) re-dissolution of secondary salts (molar ratio  $\sim 1$ ). The  $\delta^{34}\text{S}$   $-\text{SO}_4 = +19.4\text{‰} > \delta^{34}\text{S} - \text{S}$  of rock, accompanied by increased sulphate concentration also indicates an input from re-dissolution of secondary salts. Overall,  $\delta^{18}\text{O}$  composition of these non-snowpack sulphate ( $-11.9$  to  $-2.2\text{‰}$ ) mostly stayed below the threshold  $\delta^{18}\text{O}$  value ( $-6.7$  to  $-3.3\text{‰}$ ) for minimum  $\text{O}_2$  condition, suggesting that certain proportion of sulphate was regularly supplied from anaerobic sulphide oxidation.

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## 1. Introduction

Sulphate strongly influences the composition of water, conductivity and pH because many of the High Arctic glacial streams carry sulphate more than normally found in other terrestrial streams (Chillrud et al., 1994; Hindshaw et al., 2016; Tranter et al., 1993). Several hydrogeochemical studies have been conducted to understand the sources and associated biogeochemical processes (Bottrell and Tranter, 2002; Cooper et al., 2002; Hodson and Tranter, 1999; Hodson et al., 2015; Sharp et al., 1995; Telling et al., 2015; Wadham et al., 2004, 2007). Most of these studies suggest that oxidation of pyrite minerals from sedimentary deposits is the major source of dissolved sulphate. Pyrite oxidation is an exothermal process and can occur with or without microbiological activity. Separating the biotic and abiotic sulphate from natural samples is extremely difficult because of their chemical and isotopic composition. Nevertheless the rate of biologically assisted

sulphide oxidation (biotic) is generally one or two orders faster than simple chemical oxidation (abiotic) (Chen and Morris, 1972). Sulphide oxidation takes place in both, aerobic and anaerobic (Equations (1) and (2)) glacial environments (Bottrell and Tranter, 2002; Wadham et al., 2004; Wynn et al., 2006).



The premise of the sulphate-oxygen isotope synthesis depends on the stoichiometry of oxygen incorporation and the altogether different isotopic signature of both water and air  $\text{O}_2$ . Under anaerobic conditions, 100% of the oxygen is sourced from the encompassing water, whereas, under aerobic conditions, a proportion is also sourced from atmospheric  $\text{O}_2$ . Notwithstanding, fast oxygen isotopic exchange between intermediate oxidation products and the encompassing water imply that anything above 25% atmospheric  $\text{O}_2$  content reflects an oxidizing situation (Holt et al., 1981; Holt and Kumar, 1991). The percentage of atmospheric  $\text{O}_2$  finally present in the sulphate molecule depends on environmental conditions (e.g. pH and microbiological activity) which influence

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the rate of isotopic exchange (Bottrell and Tranter, 2002). This theory has successfully been applied to study Arctic glacial environment (Bottrell and Tranter, 2002; Wynn et al., 2006).



The presence and activities of sulphate reducers are also reported from the anaerobic fringes of glacial microenvironments (Skidmore et al., 2005; Wadham et al., 2004; Wolicka et al., 2014). Sulphate reduction is a multi-step process: the first step of which is endergonic hence, requires a minimum external energy supply to start the reaction. That is normally provided by some specialised bacteria (sulphate reducing bacteria) that use sulphate as a terminal electron acceptor to decompose organic substrates. Organic sources in the High Arctic glacial environment are often limited and mostly supplied by microbial biomass recycling, defoliation of sedimentary deposits and over-ridden fossil soils (Barker et al., 2006; Hood et al., 2005, 2009; Punkari and Forsstrom, 1995; Zeng, 2007). These organic matter may range from simple soluble molecules to least degradable recalcitrant forms. Generally simple soluble molecules are considered the primary choice among the organic substrate, but, the release of methane from deglaciated tills have often been connected to the degradation of recalcitrant fossil organic matter (Bardgett et al., 2007; Simpkins and Parkins, 1993). In addition to organic matter and temperature, availability of other more energy efficient electron acceptors (e.g.  $\text{O}_2$ ,  $\text{NO}_3$ ) also controls sulphate reduction activity in various environments. Thus, in an anaerobic environment, sulphate reduction controls the methanogenesis (fermentation) by providing an alternative more energy efficient flowpath for organic matter decomposition. The global significance of this control can be drawn from the fact that increased anthropogenic sulphate emissions have reduced the natural wetland methane production by 5 terragrams below pre-industrial levels (Shimel, 2004).

Previous hydrogeochemical and stable isotope works done on the Midtre Lovénbreen glacier and its catchment were largely focussed on the atmospheric nitrogen input induced biogeochemical responses of this ecosystem (Ansari et al., 2013; Cameron et al., 2012; Hodson et al., 2005, 2008, 2010; Roberts et al., 2010; Telling et al., 2012; Wynn et al., 2006, 2007). That led to an improved understanding of various nitrogen biogeochemical processing such as mineralisation, assimilation, nitrification, denitrification and nitrogen fixation (Ansari et al., 2013; Hodson et al., 2005, 2008, 2010; Telling et al., 2012; Wynn et al., 2006, 2007) in supraglacial cryoconite holes, subglacial and proglacial environments. Processes of sulphide oxidation and sulphate reduction have also been extensively addressed in this environment (Hodson et al., 2005; Wynn et al., 2006; Borin et al., 2010; Irvine-Fynn and Hodson, 2010). Like in any other Arctic glacier environment sulphide oxidation constitutes a dominant fraction of the high sulphate load commonly seen in Midtre Lovénbreen subglacial or in proglacial runoff. Sulphate concentrations in the Midtre Lovénbreen subglacial runoff are generally two times higher than that achievable with consumption of dissolved oxygen (Tranter et al., 2002; Wynn et al., 2006; Irvine-Fynn and Hodson, 2010). Therefore the role of bacterial catalysis and additional oxidants (mainly  $\text{Fe}^{3+}$ ) are commonly assumed as the most likely source of the excess sulphate. Anaerobic processes like denitrification and sulphate reduction in Midtre Lovénbreen glacier and the nearby area, have been mostly found associated with lowflow/delayed flow subglacial runoff (Hodson et al., 2005; Ansari et al., 2013; Wynn et al., 2006). Nevertheless the spatial dynamics of aerobic and anaerobic sulphide oxidation processes and re-dissolution of secondary salts to the bulk sulphate loading in subglacial runoff and proglacial streams flowing away from the

glaciers are yet to be determined. Therefore, this study measured downstream changes in stable isotopic compositions and concentrations of sulphate in the two major streams to comprehend the redox variability and sources of sulphate over the subglacial and proglacial flowpaths.

## 2. Field site and methodology

### 2.1. Midtre Lovénbreen

Midtre Lovénbreen (78.53°N and 12.04°E) is a 6 km long glacier which occupies an area of approximately 5.5 km<sup>2</sup>, established on the Brøggerhalvøya peninsula in North West Spitsbergen (Fig. 1). Its height ranges from 50 to 600 m above sea level and maximum depth of this glacier is approximately 180 m. Due to high ice overburden, a large portion of the glacier bed is at the pressure melting point and therefore maintains a wet subglacial condition whole year. During winter, some subglacial meltwater gets stored until a subglacial upwelling emerges near the glacier snout (usually in the month of July). Through the summer the upwelling increasingly discharges fresh meltwater supplied from the high elevation crevasses, rather than englacial and subglacial ice-melt. In addition, Midtre Lovénbreen catchment is characterised by quaternary moraines composed of bedrock sourced from the surrounding topography (Hjelle, 1993).

### 2.2. Sampling sites and sampling frequency

Winter deposition sampling: five bulk snowpack samples (SG1 to SG5 see in Fig. 1) were collected from the centre line of the glacier surface at approximately 80–90 m elevation intervals on DOY 160 during 2009 and DOY 143–149 during 2010. These samples were collected using a pre-cleaned HDPE tube of known dimensions. The tube was first cleaned at least three times with surface snow from near the site, then it was inserted vertically into the snowpack up to the glacial surface and excavated using a clean knife. The snow cores were separately transferred into pre-cleaned polyethylene zip-lock bags (three times washed with snow) and weighed.

Stream samples were collected from DOY 184 at MLE and MLW, and from DOY 198 at MLU, following subglacial upwelling emergence (Fig. 1). During 2009, meltwater samples were collected every second or third day from DOY 184 to 205 and then sampling recommenced from DOY 233 to 250 after an intervening period of rain and melt-induced high flows between DOY 206 and 231. Initially MLE received a discharge mostly from supraglacial and glacial marginal ice melts, however, following the emergence of subglacial upwelling on DOY 196 MLE became dominated by MLU discharge.

During 2010, sampling was designed to investigate the downstream biogeochemical changes in the streams using three further sampling sites (MLW1, MLW2, MLW3) on the western stream and 4 sites (MLU, MLE1, MLE2, MLE3) on the eastern stream (Fig. 1), beginning on DOY 186. The five transects were sampled weekly from the above stream sites throughout the summer until DOY 250. This time subglacial upwelling emerged before the start of stream sampling, therefore discharges at MLE1 and MLE2 were dominated by subglacial upwelling (MLU) water while discharge at the upstream site MLE3 was predominately derived from glacial marginal ice melting.

All the samples were filtered using 0.45 µm pore size cellulose nitrate filter papers at the British NERC Arctic station lab facility (1–2 h after sample collection). Then samples were transferred into 60 mL polyethylene bottles and refrozen at –20 °C stored and frozen in polypropylene bottles and transported to the laboratory

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