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Glacier ecosystem response to episodic nitrogen enrichment in Svalbard, European High Arctic

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Abstract We describe the climatology, hydrology and biogeochemistry of an extreme nitrogen deposition event that occurred in the highly glacierised environment of the European High Arctic during June 1999. Meteorological analysis, three-dimensional air mass trajectories and a 3D transport model show that blocking high pressures over Scandinavia and the rapid advection of western European pollution toward Svalbard were sufficient to cause the most concentrated (1.15 ppm NO_3–N and 1.20 ppm NH_4–N), high magnitude (total 26 mm and up to 2.4 mm h^{-1} at 30 m above sea level) nitrogen deposition event on record in this sensitive, high Arctic environment (78.91° N, 11.93° E). Since the event occurred when much of the catchment remained frozen or under snow cover, microbial utilisation of nitrogen within snowpacks and perennially unfrozen subglacial sediments, rather than soils, were mostly responsible for reducing N export. The rainfall event occurred long before the annual subglacial outburst flood and so prolonged (ca. 10 day) water storage at the glacier bed further enhanced the microbial assimilation. When the subglacial outburst eventually occurred, high runoff and concentrations of NO_3^- (but not NH_4^+) returned in the downstream rivers. Assimilation accounted for between 53 and 72% of the total inorganic nitrogen deposited during the event, but the annual NO_3^- and NH_4^+ runoff yields were still enhanced by up to 5 and 40 times respectively. Episodic atmospheric inputs of reactive nitrogen can therefore directly influence the biogeochemical functioning of High Arctic catchments, even when microbial activity takes place beneath a glacier at a time when terrestrial soil ecosystems remain frozen and unresponsive.

Keywords Nitrogen enrichment · Arctic ecosystems · Arctic glaciers · Meltwater biogeochemistry

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Introduction

Ecosystems in Svalbard of the European High Arctic receive acidic inputs from polluted, low latitude sources following tropospheric transport (e.g. Barrie 1986; Law and Stohl 2007; Stohl 2006). Deposition of the pollution results in an anthropogenic signal in Svalbard snow (Gjessing 1977), with layers showing enhanced concentrations of NO_3^-, SO_4^{2-}, and NH_4^+ that greatly influence pH (Simoes and Zagorodnov 2001). The acidic inputs are readily observable in the

last 150 years of Svalbard's ice core record, with some decreases in recent years that may reflect stricter emission controls (e.g. Isaksson et al. 2001). Atmospheric pollution is monitored in Ny Ålesund, Svalbard, and so several important studies have examined its transfer to this site (e.g. Beine et al. 1996; Eneroth et al. 2003). The 10-year climatology of long-range transport to Ny Ålesund that was presented by Eneroth et al. (2003) identified eight major transport patterns to this part of Svalbard, including enhanced winter transport of polluted air from both Europe and Russia.

The transport of pollution to Svalbard during summer seems less common than winter (Eneroth et al. 2003), but episodic pollution events during this period of increased biological activity do occur (e.g. Krawczyk et al. 2008). The episodic delivery of both NO_3^- and NH_4^+ during summer therefore means that productivity-limiting nutrients are delivered suddenly to Arctic ecosystems that are known to be severely nutrient-limited (Shaver and Chapin 1980). To date, however, studies have largely concentrated upon the impacts of either increased availability of multiple nutrients such as N, P and K to predict impacts of greater mineralisation after soil warming (e.g. Wookey et al. 1993; Chapin et al. 1995; Schmidt et al. 1999), or upon long-term chronic N loading from the atmosphere (Gordon et al. 2001). Thus little consideration has been given to how different timing, magnitude and frequency scenarios for nitrogen deposition might influence ecosystem response. Critically, little attention has been given to short-lived episodes of nitrogen enrichment and its influence on ecosystem dynamics in the High Arctic (e.g. Tye et al. 2005).

Streamflow nitrogen dynamics during snowmelt are often used to characterise the response of watershed biogeochemical cycles to atmospheric deposition (e.g. Brooks et al. 1998; Campbell et al. 2000; Sickman et al. 2001; Tranter 1991; Petrone et al. 2007). These dynamics are, however, rather poorly understood in environments dominated by permafrost and extensive glacier ice cover, as is the case in Svalbard. Here, the biogeochemical cycling of atmospheric nitrogen can be surprisingly important, as the following aspects of recent research testify:

- Assimilation of NO_3^- and NH_4^+ occurs in glacial snowpacks almost as soon as they become isothermal during the summer (Hodson 2006; Hodson et al. 2005a). Assimilation also occurs after the retreat of the transient snow line due to primary and heterotrophic microbial production within small melt pools on the glacier surface known as "cryoconite holes" (e.g. Stibal et al. 2006). However, since cyanobacteria are present, NO_3^- assimilation might be offset by N_2 fixation.
- Biological nutrient demands in cryoconite holes (1–10% of the ice surface) and wet snowpacks (up to 100% of the ice surface) can account for much of the annual NH_4^+ deposition upon Midtre Lovénbreen, Svalbard (the site of the present study: Säwström et al. 2002; Hodson et al. 2005a, 2007; Anesio et al. 2009).
- Surface melt can supply water and NO_3^- to the glacier bed and promote denitrification in zones of slow transfer through hydraulically inefficient sediments. At Midtre Lovénbreen, the process imparts a significant impact upon the $\delta^{15}N$ and $\delta^{18}O$ signatures of NO_3^- early in the summer (Wynn et al. 2006).
- Stable isotope observations also show that NO_3^- production is more important than denitrification when flow rates through glacial sediments increase (Wynn et al. 2007). As a result nitrification might be the most obvious microbial control upon meltwater NO_3^- dynamics during the summer, whilst denitrification is more pronounced during winter, spring and early summer (Hodson 2006, Wynn et al. 2006, 2007). These dynamics are not exclusive to the subglacial environment at Midtre Lovénbreen, because they are also apparent in lateral talus and moraine waters sampled at the margin of a cold-based Antarctic glacier (Hodson 2006; Hodson et al. 2009).

The above characteristics therefore suggest that both the timing and magnitude of atmospheric nitrogen loading events could be critical for understanding catchment biogeochemical response. The purpose of this paper is therefore to examine the causes, origins and impact of a high magnitude NO_3^- and NH_4^+ deposition episode that occurred during early summer in June 1999. This rain-on-snow event took place prior to the development of the active layer in lower elevation catchment soils and so it allows us to emphasise the biogeochemistry of

reactive nitrogen transfer through snowpack and glacial flowpaths instead.

Field site

The study was conducted in Ny Ålesund, Svalbard, where the Zeppelin Atmospheric Monitoring and Research Station overlooks Midtre Lovénbreen. This glacier is ca. 5.4 km^2 and occupies a ca. 9.4 km^2 catchment (Fig. 1). Atmospheric monitoring activities at the Zeppelin Station are numerous and include gaseous and aerosol-phase characterisations (see below), whilst precipitation is measured in Ny Ålesund. These data contribute to the European Monitoring and Evaluation Programme (EMEP, http://www.emep.int).

The hydrology and biogeochemistry of the Midtre Lovénbreen catchment are well known and have been studied intensively since 1997 (see Hodson et al. 2000, 2005a, 2007; Wadham et al. 2006; Wynn et al. 2006, 2007). Water balance calculations for the period 1997–2002 show that summer precipitation (usually rain) typically delivers 0.49 (±0.12) m a^{-1} and therefore makes a significant contribution to specific annual runoff (1.1–1.5 m a^{-1}: see Hodson et al. 2005b). Runoff is carried by two major streams that drain the lateral flanks of the glacier (MLE and MLW: Fig. 1). Additionally, between one quarter and one-third of the runoff is routed through the subglacial reservoir which discharges (MLSG) into one of these streams following a subglacial outburst flood that usually occurs in the first half of July (Hodson et al. 2005b).

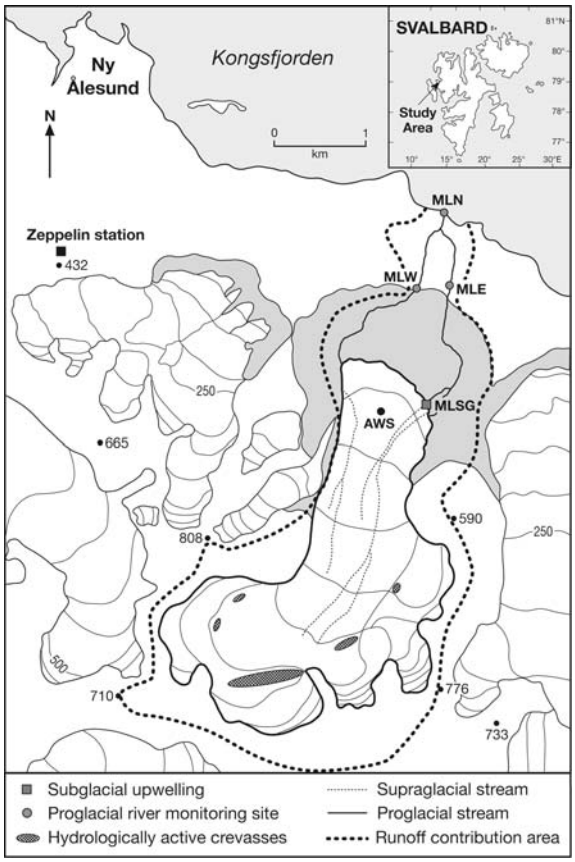

Fig. 1 The study site and the sampling points within it

Methods

Hydrology and hydrochemistry

Full details of the discharge and water chemistry measurements during the observation period at Midtre Lovénbreen (i.e. Day of Year (DOY) 169–210, or 18th June–29th July, 1999) are reported elsewhere (Hodson et al. 2005a, b; Mumford 2002). Briefly, major anions were determined using a Dionex DX-100 ion chromatograph; NO_3^- using the manual cadmium reduction method (Mackereth et al. 1978) and NH_4^+ using a FOSS-Tecator FIAstar 5000 following conversion to NH_3 by NaOH addition (FOSS-Tecator 2000). Precision errors were 2.6% for NO_3^- and 5.1% for NH_4^+. Major anions (Cl^- and SO_4^{2-}) were quantified using a Dionex DX100 ion chromatograph with precision errors $\leq$5% (Hodson et al. 2005a). Samples were collected from various sites in the catchment (Fig. 1), including the subglacial drainage site (MLSG), a bulk meltwater site adjacent to the fjord (MLN) and the two lateral streams described above (MLW and MLE).

The flow measurements involved the logging of 30 s pressure records at MLE and MLW (see Fig. 1). These data were averaged and stored as hourly values on a Campbell Cr10x data logger. Calibration of the pressure transducer records was undertaken using daily streamflow velocity area measurements. Errors in the discharge record were typically 10% and full details are reported in Hodson et al. (2005a, b).

Meteorology and atmospheric chemistry measurements

A broad meteorological characterisation of the conditions during the June 1999 event was performed with data from Deutche Wetterdienst (www.dwd.de) and satellite images from Dundee University (www.sat.dundee.ac.uk). Additionally, meteorological data for the Midtre Lovénbreen catchment were collected using a Campbell Scientific automatic weather station installed upon the glacier at ca. 75 m elevation (see Hodson et al. 2005b). Precipitation quantity was measured on a weekly basis within Ny Ålesund (see below) and on an hourly basis within Midtre Lovénbreen's forefield at MLE (Fig. 1). Figure 2 shows the relationship between these two low elevation rainfall gauges after aggregating hourly measurements in order to match the weekly resolution of the Ny Ålesund data. Precipitation recorded at MLE typically exceeded that recorded in Ny Ålesund by a factor of ca. 30% over the 1999 and 2000 summers (Fig. 2).

Inorganic trace gases and aerosol particles were sampled on a 24-h basis at Zeppelin Station by the Norwegian Institute for Air Research (NILU) and are used here to characterise nitrogen abundance in the air masses being advected over Midtre Lovénbreen. NH_3 and HNO_3 gases, plus the water-soluble NO_3^- and NH_4^+ components of aerosol particles were analysed using ion chromatography. Full details are available from the EMEP and NILU website (http://tarantula.nilu.no/projects/ccc/). The precipitation monitoring, which is also overseen by NILU, was undertaken in Ny Ålesund, rather than at the mountain station. Quasi-weekly samples from a bulk deposition collector were analysed for pH and then for ionic constituents using liquid ion chromatography. The bulk sampler material and design conform to EMEP standards (see web link above for further details), the balance between wet and dry deposition in the sampler is discussed in conjunction with our field data.

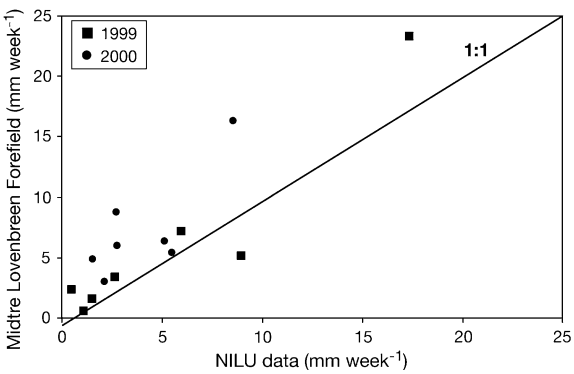

Fig. 2 Comparison between rainfall receipt at Ny Ålesund and in Midtre Lovénbreen forefield (30 m above sea level) during the summers of 1999 and 2000

Trajectory modelling

Three-dimensional, 5-day back trajectories were calculated using the McGrath (1989) model and wind fields from the European Centre for Medium-Range Weather Forecasts (ECMWF) operational model. The fields have a horizontal resolution of 1° × 1° and a temporal resolution of 6 h. Linear interpolation was therefore performed to deduce the wind velocities at each point along the trajectories at a 1-h resolution. Trajectories arriving at Ny-Ålesund (79.9° N, 11.9° E) at 750, 850 and 950 hPa were calculated twice daily (00UTC and 12UTC), for the period June 23, 1999 to June 30, 1999. For simplicity, we present just the trajectories arriving at Zeppelin mountain at 850 hPa, as they are elevated high enough to limit the surface effects in the wind data. Trajectories for adjacent nodes in the grid are not shown here as they indicate the same broad air mass trajectories.

Transport model

We used MATCH, a eulerian three-dimensional atmospheric diffusion model, to explore the capacity for prevailing meteorological conditions to deliver nitrogen to the study site from European industrial and natural sources. The MATCH model (Multiple-Scale Atmospheric Transport and Chemistry modelling system: Engardt et al. 1998; Robertson and Langner 1998) has been developed as a flexible, offline chemistry/transport/deposition model for atmospheric pollutants. It is used in a range of applications from urban-scale studies on 1 km or higher horizontal resolutions, to continental-scale studies on acid deposition and photochemistry (Langner et al. 2005). The model reads in modules of parameterisations for emissions, vertical distributions, deposition processes and chemical processes, which in the present study, included reactions and transformations of ten components of ammonia, SO_x

and NO_x in the manner described by Hov et al. (1994). Data characterising emissions of ammonia, SO_x and NO_x and also boundary conditions were taken from the Emission Database for Global Atmospheric Research (EDGAR) data centre from 1995 (http://arch.rivm.nl/env/int/coredata/edgar). These global fields include both anthropogenic and natural sources with a resolution of 1° × 1°. Emission and boundary fields were read in with the same 6 hourly time resolution as the meteorological fields and transformed to hourly data using linear interpolation. Dry deposition was modelled using a resistance approach, whilst wet scavenging was set to be proportional to the precipitation intensity using species-specific scavenging coefficients. Expressions for the photolysis rates, depending on solar elevation derived for clear sky situations, were taken from Derwent and Jenkin (1990). To account for the effect of clouds, the photolysis rates were scaled by the ratio of the actual global radiation to the clear sky global radiation. The results of this modelling are reported in the present paper as boundary layer NO_y profiles along the air parcel trajectory. Since the modelling was conducted without full characterisation of the phases and reactions involved, the profiles are only intended to provide a qualitative basis for interpreting NO_3^- and NH_4^+ delivery to the field site. Further details of the MATCH model and its application here are available from Julin (2003).

Nitrogen mass balance

Hodson et al. (2005a) have already considered nitrogen mass balance in the Midtre Lovénbreen catchment during the 1998/1999 and 1999/2000 water years. However, the event during June 1999 was poorly constrained within this mass balance scheme because the emphasis was upon annual retention and production of nitrogen, not the fate of a short-lived event. We therefore resorted to a new scheme in the present study. First, we estimated atmospheric deposition of NO_3^- and NH_4^+ during the observation period (i.e. DOY 169–210) from the product of the NILU concentration data set (Table 1) and our own precipitation quantity estimates. Since there is uncertainty in the distribution of precipitation events across mountainous environments, our rainfall gauge data from MLE were distributed over the catchment in 50 m elevation zones using three different precipitation gradients (i.e. $\Delta Ppt^{1,2,3}$). For ΔPpt^1, no elevation change in precipitation amount was assumed (i.e. $\Delta Ppt^1 = 0\%/50$ m). For ΔPpt^2, a 10% increment per 50 m was applied, but only up to 500 m ($\Delta Ppt^2 = 10\%/50\ m^{<500}$). ΔPpt^2 was based upon the only empirical relationship established between elevation and precipitation in the Ny Ålesund area (see Førland et al. 1997), which implied a clear altitudinal limit to the gradient at 500 m. Finally, ΔPpt^3 assumed an empirical increase based upon average winter snow accumulation (see Hodson et al. 2005a, b). The ratio of total catchment rainfall estimated using ΔPpt^1, ΔPpt^2 and ΔPpt^3 to rainfall measured at the gauge was 1, 1.8 and 3.0 respectively (Table 1).

Second, daily fluxes of total NO_3^- and NH_4^+ transported by the MLE, MLW and MLN streams were estimated for the observation period (DOY 169–210) from the product of the daily concentration values and the runoff volume. At MLN, our sampling frequency had a 2 day period and so linear interpolation between successive values was used to produce a daily concentration series. Runoff was estimated here as the sum of MLE and MLW runoff data, because visual observations and ca. ten velocity-area estimates of discharge at MLN showed no significant sources/sinks of water between the sites concerned.

Third, we estimated the daily stream concentrations of event-derived NO_3^- and NH_4^+ (hereafter $^{event}NO_3^-$ and $^{event}NH_4^+$) at MLE, MLW and MLN by subtracting all other NO_3^- and NH_4^+ sources from the total concentration. Concentrations of

Table 1 Characteristics of the quantity and quality (NILU data) of the precipitation event, as compared to long-term (1996–2003) volume-weighted mean (VWM) concentrations

Precipitation in forefield (mm)	25.7
Total catchment estimate 1 (ΔPpt^1)	23.3
Total catchment estimate 2 (ΔPpt^2)	46.3
Total catchment estimate 3 (ΔPpt^3)	77.1
pH	4.2, 3.2 (4.1)
NO_3^- (μeq L^{-1}, (ppm N))	82.1 (1.15)
NH_4^+ (μeq L^{-1}, (ppm N))	85.7 (1.20)
VWM pH*	6.51
VWM NO_3^+ (μeq L^{-1}, (ppm N))	9.14 (0.128)
VWM NH_4^+ (μeq L^{-1}, (ppm N))	19.1 (0.267)

The pH data are for two successive aliquots that were analysed, whilst the value in parenthesis is the volume-weighted average pH of these aliquots. The acidic aliquot was too small for NO_3^- and NH_4^+ analysis. ΔPpt^1, ΔPpt^2, ΔPpt^3 are the three precipitation gradients used

$^{event}NO_3^-$ and $^{event}NH_4^+$ were then multiplied by daily runoff volumes to estimate stream fluxes of $^{event}NO_3^-$ and $^{event}NH_4^+$ throughout the summer. Since Cl^- concentrations in all summer deposition events varied little and were similar to those in the winter snowpack, $^{event}NO_3^-$ and $^{event}NH_4^+$ concentrations were estimated using:

$$^{event}NO_3^- = NO_3^- - aCl^- \quad (1)$$

$$^{event}NH_4^+ = NH_4^+ - bCl^- \quad (2)$$

where a and b are the pre-event (DOY 169–176), average NO_3^-/Cl^- and NH_4^+/Cl^- equivalent concentration ratios respectively. The values for a and b were 0.086 and 0.038 for both MLE and MLW, and 0.045 and 0.020 for MLN. Strictly speaking, the quantities aCl^- and bCl^- account for the net release of inorganic nitrogen after the leaching of debris (moraine, talus, organic matter) and snow (i.e. snowpack elution) by a mixture of meltwater and the light, unpolluted rainfall that occurred later on in the summer (Hodson et al. 2005a; Wadham et al. 2006). Since the a and b coefficients were estimated using the very first (snowmelt) runoff samples that were collected, $^{event}NO_3^-$ and $^{event}NH_4^+$ were probably underestimated later in the summer when less snowcover existed. However, the likely errors in the mass balance estimates are insignificant because the event occurred so early in the summer.

Lastly, in order to account for the minor fluxes of event nitrogen transferred by the MLE, MLW and MLN rivers after we had finished monitoring, concentrations of $^{event}NO_3^-$, and $^{event}NH_4^+$ were assumed to decay to 0 ppm N by the end of the summer in a linear fashion. It was therefore assumed that all of the freely available June 1999 event nitrogen had been removed from the system by DOY 274. The missing daily runoff volumes were then estimated using regression models that used full discharge records from an adjacent catchment as the predictor (see Hodson et al. 2005a, b for details).

Results

Hydrometeorology

Figure 3a and b depict the hydrometeorological conditions recorded in the Midtre Lovénbreen basin during summer 1999. This shows high flows at MLE on DOYs 173-175 due to snowbank collapse (and a significant error in the runoff estimates we suspect) and then increasing flows at both MLE and MLW in direct response to the event under study. Rainfall first fell in the afternoon of DOY 175 as an event of 9.95 mm that persisted for 1 day. Then a small event on DOY 177 (just 1.40 mm) took place, followed by 14.33 mm of precipitation during DOY 178 and 179. It was almost certainly this event which caused the nitrogen deposition episode, because precipitation intensities were high early on (up to 2.5 mm h^{-1}) allowing an immediate response to be discernable from the chemistry of our daily stream samples. A small volume of precipitation was also recorded after this period on DOY 180 (Fig. 3a) and sampled separately by NILU. Our records show that this was the final part of the same precipitation event. Due to the sampling intervals used by NILU, the chemical characterisation of the event clearly lacks the resolution of the hourly precipitation quantity data shown on Fig. 3a. We therefore define the precipitation "event" of interest as the 26 mm of rain that was captured in the glacier forefield between DOY 175–181: a total amount that most likely represented between 46 and 77 mm of precipitation across the entire catchment (Table 1). Interestingly, its impact upon streamflow, though obvious, only generated seasonal maximum flows at MLW because even higher flows occurred at MLE (and MLN downstream) in response to seasonal maximum air temperatures between DOY 198 and 203. The seasonal maximum flows that occurred later at MLE and MLN were also associated with the outburst of water from beneath the glacier via MLSG. These outburst waters were not routed into the MLW stream.

Chemistry

NO_3^- and NH_4^+ concentrations in air, precipitation and total catchment runoff (Site MLN in Fig. 1) are shown in Fig. 4a and b respectively. The air measurements show small increases coincident with the event under study around Day 179, whilst the precipitation and stream records show prominent maxima for NO_3^- and NH_4^+ concentrations. Maximum stream concentrations of NO_3^- and NH_4^+ at MLN were ca. 25 and 17% respectively of the precipitation concentrations during the event. The

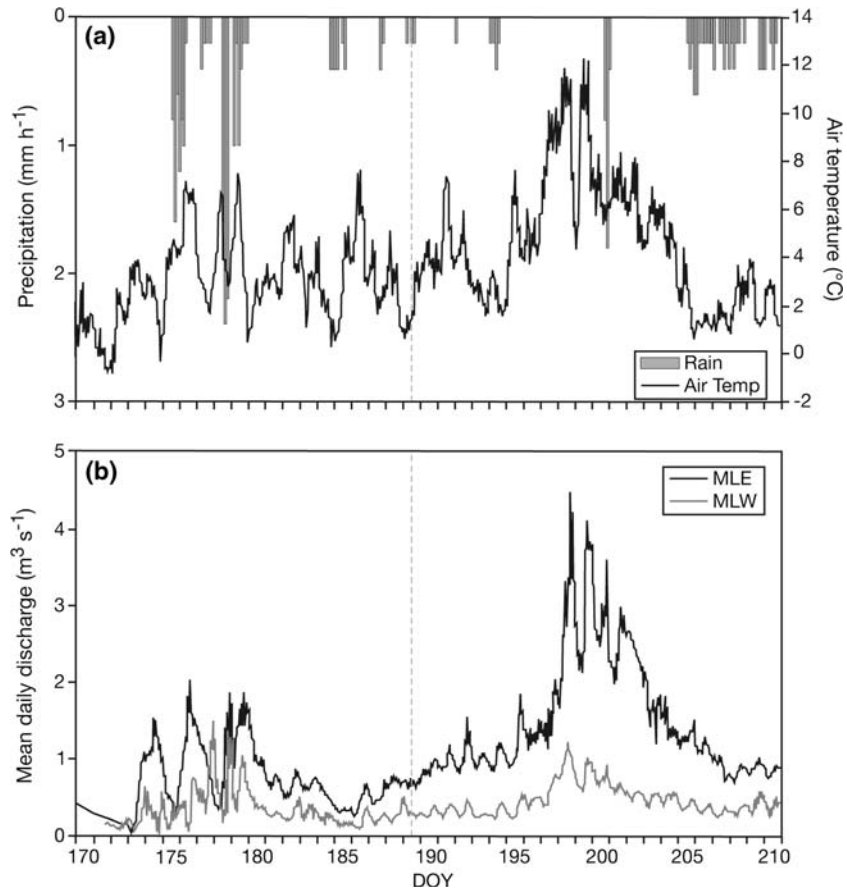

Fig. 3 Hydrometeorological conditions during the observation period of summer 1999. **a** Precipitation and air temperature. **b** Runoff at MLE and MLW

seasonal maximum air concentrations of both NO_3^- and NH_4^+ occurred later in the summer of 1999, and reflect the advection of pollution from Europe via eastern Russia: a common mode of air flow toward Svalbard (Eneroth et al. 2003). However, neither of these maxima in air concentrations forced a significant increase in stream concentrations, suggesting that deposition rates remained low. We therefore conclude that the June 1999 event occurred primarily via wet deposition, with a composition dominated by NO_3^-, NH_4^+ and SO_4^{2-} (SO_4^{2-} data not shown) that is characteristic of anthropogenic pollution.

Figure 5a–d present the molar concentrations of Cl^-, NO_3^-, NH_4^+ and pH respectively for the four stream sampling sites across the catchment. Prior to the event (i.e. up to DOY 177), concentrations of both Cl^- and NO_3^- were declining due to a snowpack ion elution process (Fig. 5a, b). Usually, elution means that low concentrations typify late summer runoff, with increasingly dilute snowmelt being further diluted by icemelt runoff (e.g. Tranter et al. 1996). It is interesting however that NH_4^+ does not show any evidence of elution early in the runoff period in Fig. 5c. In fact, apart from when stream flow chemistry is clearly affected by the precipitation event (Day 179–191), NH_4^+ concentrations are nearly always close to detection and lower than snowpack concentrations (Hodson et al. 2005a).

Figure 5b and c also show that all streams responded to the June 1999 event by displaying markedly higher NO_3^- and NH_4^+ concentrations on Day 179 and thereafter. Cl^- concentrations did not increase (Fig. 5a), largely because Cl^- abundance in the rainfall was very similar to that in the streams prior to its occurrence (i.e. 2.55 ppm). Later on in the summer, it is also notable that the subglacial outburst

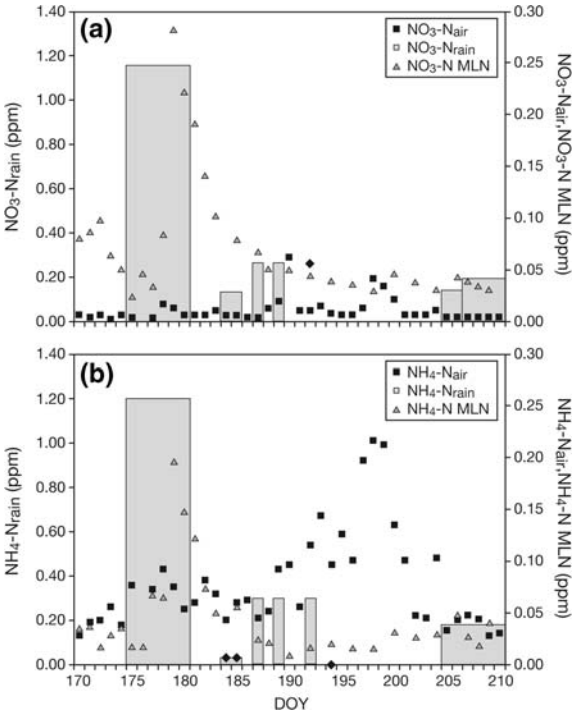

Fig. 4 Chemical conditions during the 1999 observation period: 1. **a** NO_3–N in air (ppm), runoff at MLN and rain (both ppm). **b** NH_3–N in air (ppm) and NH_4–N in rain and runoff at MLN (ppm)

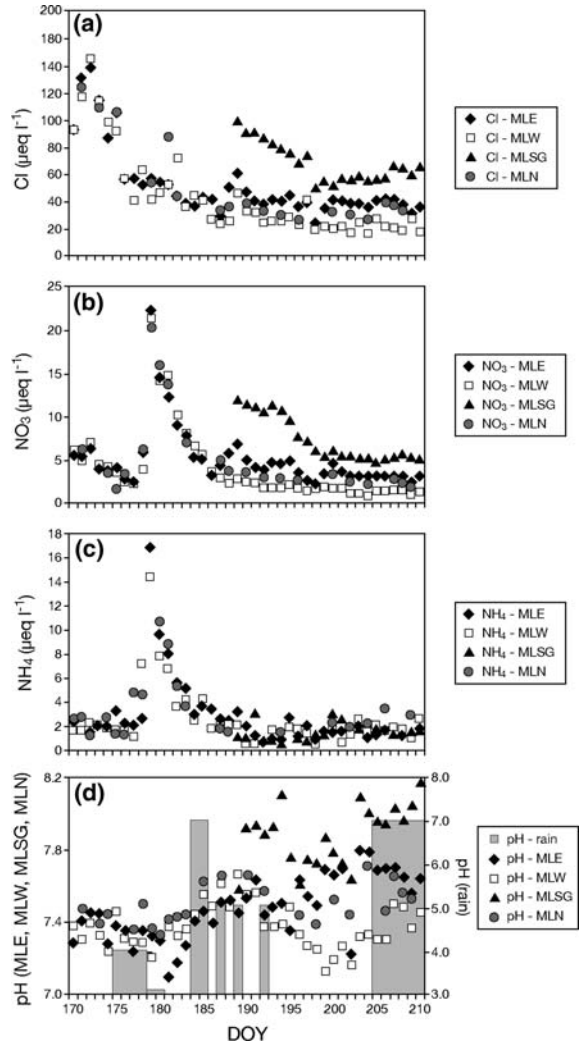

Fig. 5 Concentrations of **a** Cl^-, **b** NO_3^-, **c** NH_4^+ and **d** pH for the streams MLE, MLW, MLN and MLSG. Samples from MLSG begin following a subglacial outburst event on DOY 188

into MLE and MLN (starting on DOY 189) temporarily offset the declining solute concentrations. This is because the subglacial drainage waters that outburst into the proglacial rivers upstream of MLE included a mixture of early season snowmelt and precipitation event waters. The June 1999 event waters therefore re-emerged from the glacier after this outburst took place and caused concentrations of most solutes and pH to increase at MLE (and to a lesser extent MLN) relative to MLW. Again though, it is notable that NH_4^+ concentrations did not increase following this outburst.

Figure 5d shows that the pH of the precipitation event was measured in two aliquots: an initial, large aliquot that was sufficient for all analyses (i.e. the DOY 175–179 events) and a second aliquot that fell into the next sampling interval but was of insufficient volume for a complete chemical characterisation (i.e. the DOY 180 sample). Table 1 shows that the corresponding pH values are different: being 4.21 for the main event water and 3.18 for the second minor aliquot. The latter result, if accurate, represents the most acidic precipitation event yet recorded at Ny Ålesund. However, given the very small liquid volumes involved, we suggest that it might instead be an artefact of the very small precipitation volume in the collector. The value for the initial, larger aliquot is therefore more likely to be representative of the event, and is significantly more acidic than the long-term, volume-weighted mean for Ny Ålesund of 6.51 (Table 1). However, there appears to be no significant depression in the pH of the streams draining the catchment, with only MLE showing a decline to a seasonal minimum pH of 7.1 during the event.

Mass balance

Table 2 shows the mass balance calculations after estimating inputs and outputs of $^{event}NO_3^-$ and $^{event}NH_4^+$ using the three different precipitation gradients and the stream fluxes. All calculations indicate a runoff deficit in both forms of nitrogen within the catchment, but to varying degrees. Thus ΔPpt^1 produces a near balance (i.e. minor deficit) of $^{event}NO_3^-$, yet a substantial $^{event}NH_4^+$ deficit. ΔPpt^3 indicates a significant deficit in both $^{event}NO_3^-$ (67%) and $^{event}NH_4^+$ (77%). It should be noted that virtually none of this deficit can be accounted for by retention within a residual snowpack due to the near absence of snow cover by the end of the summer (see Hodson et al. 2005a).

In all cases the NH_4^+ deficit appears to have been more important than the NO_3^- deficit. Thus, while the loading of NO_3^- and NH_4^+ into the catchment during the event were markedly similar in magnitude, the $^{event}NO_3^-$ stream fluxes were far greater than $^{event}NH_4^+$ fluxes, especially for longer residence time flowpaths (i.e. MLSG).

Table 2 Estimates of nitrogen inputs, outputs and retention during the event. In^1, In^2 and In^3 indicate use of the three different precipitation gradients (ΔPpt^1, ΔPpt^2 and ΔPpt^3) respectively to estimate atmospheric inputs during the event and their contribution to total annual deposition (estimated using winter snowpack data presented by Hodson et al. (2005a)) is also shown

	$^{event}NH_4^+$	$^{event}NO_3^-$	$^{event}DIN$
Mass fluxes (kgN)			
In^1	252	252	504
In^2	444	443	887
In^3	747	746	1493
MLE riverine flux	−117	−197	−314
MLW riverine flux	−56	−51	−107
[MLE + MLW] riverine flux	−173	−248	−421
MLN riverine flux	−180	−281	−461
% of total annual deposition			
In^1	72	66	69
In^2	80	77	78
In^3	84	84	84
Runoff deficit (% of input)			
Deficit for In^1	31	2	16
Deficit for In^2	61	44	53
Deficit for In^3	77	67	72

Atmospheric modelling

Figure 6 shows the simulated time series of the vertical NOy concentration profile leading to the June 1999 event. The air mass trajectories associated with the time series are shown in Fig. 7, and they clearly link the high NO_y observed in Ny Ålesund to UK emissions. Our examination of the meteorological situation during 23–28th of June 1999 (DOY 174–179) supports this because high pressure existed over Europe and Great Britain, which ensured stable stratification and no significant mixing of the air column. This, in conjunction with low wind speeds, enabled the accumulation of high concentrations of pollutants from anthropogenic source areas. The high pressure then moved towards Scandinavia and the pressure over Great Britain decreased. The boundary layer over Great Britain then extended in altitude and the accumulated pollutants were transported northwards above the still stable stratified boundary layer over the sea. These conditions will have limited the vertical exchange with the ground/sea surface and favoured rapid transport of pollutants directly to the Arctic. Since this meteorology suppresses precipitation, little washout probably occurred en route to Svalbard. Further, the delivery of the nitrogen was also enhanced by environmental and climatic factors due to the interaction between the airflow and the mountainous topography of western Spitsbergen.

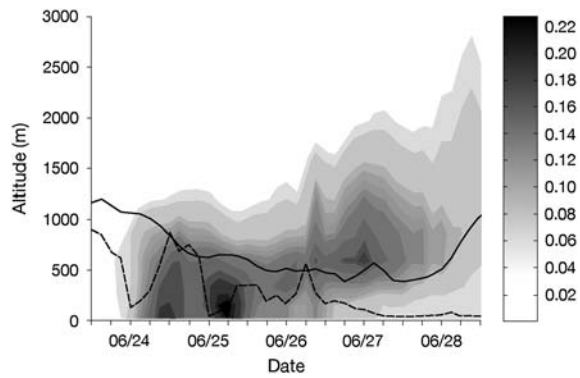

Fig. 6 Simulated vertical NO_y concentration profiles (ppmv) for the air mass trajectory arriving at the Zeppelin Station on June 28th, 1999 (DOY 179). The solid and dashed lines show the trajectory path and boundary layer, respectively

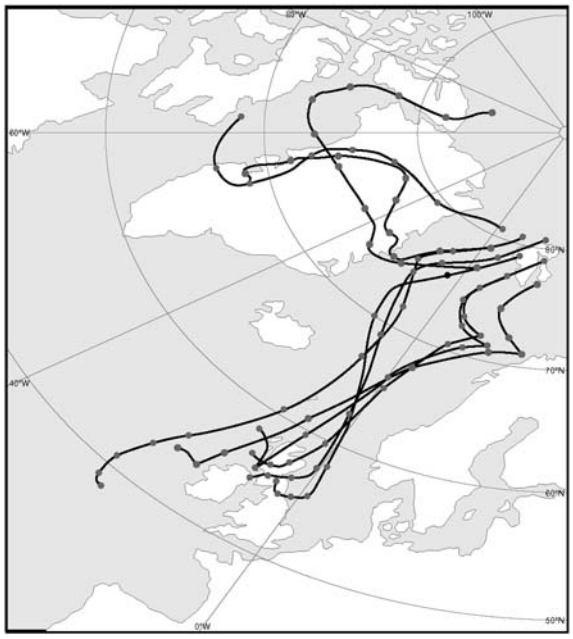

Fig. 7 Five day air mass trajectories arriving at and around the Zeppelin Station on June 28th, 1999 (1200, 850 hPa, DOY 179)

Discussion

Acid deposition upon Svalbard has been known to occur for some time (Gjessing 1977) and readily produces acid pH values in its snowpacks (Simoes and Zagorodnov 2001). These studies suggest that the most reliable estimate of the precipitation event pH during June 1999 (i.e. 4.21) is not at all atypical for Svalbard. Therefore the most striking characteristic of this event was its nitrogen load, which caused the annual wet deposition to be greatly enhanced in 1999 compared to other years (AMAP 2006). Further, Table 3 shows that the 1999 annual runoff yields of NO_3^- and NH_4^+ were up to 5 and 40 times respectively those reported in either the Midtre Lovénbreen catchment or its neighbour, the Bayelva catchment. The following discussion therefore explores the extent to which both the causes and consequences of the June 1999 event were unique, whilst also attempting to improve our understanding of the biogeochemistry of atmospheric nitrogen in transit through the glacial environment.

Atmospheric conditions responsible for the event

Figure 6 shows both the simulated atmospheric concentration of NO_y and the heights of the modelled trajectory boundary layer, revealing how ground level emissions enhanced the NO_y concentration on 24th and 25th June (DOYs 175 and 176) when the trajectory was over the UK. The modelling results also indicate that uplift of this pollution above the boundary layer then occurred, enabling its rapid transport to Ny Ålesund over the next 3 days. The simulated time-evolution of the pollution plume that reached Ny Ålesund was therefore able to increase NO_y concentrations to 0.05–0.1 ppmv above background levels, which are typically sub-ppbv. The scavenging of the NOy by precipitation in Ny Ålesund therefore resulted in the enhanced nitrogen load associated with the June event.

The broad synoptic conditions responsible for the above advection of nitrogen to the study site and shown in Fig. 7 conform to Eneroth et al.'s (2003) Cluster 7, which represents air mass transport to

Table 3 Published total NO_3^- and NH_4^+ yields for glacierised basins in the vicinity of Ny Ålesund

Year	Site	NO_3 yield (kgN km^{-2} a^{-1})	NH_4 yield (kgN km^{-2} a^{-1})	References
1991	Bayelva	13	–	Hodson et al. (2000)
1992	Bayelva	13	–	Hodson et al. (2000)
1997	Midtre Lovénbreen	25	–	Hodson et al. (2000)
1998	Midtre Lovénbreen	28	–	Hodson et al. (2000)
1999	Midtre Lovénbreen	61	40	Hodson et al. (2005)
2000	Midtre Lovénbreen	22	3.0	Hodson et al. (2005)
2000	Bayelva	24	3.3	Hodson et al. (2005)
2002	Midtre Lovénbreen	15	1.4	Wynn et al. (2007)

The Bayelva catchment is immediately west of the Midtre Lovénbreen catchment

Svalbard from the Atlantic region between Greenland and Western Europe. This is a rather large and heterogeneous source area and so variable amounts of pollution can be seen in the corresponding EMEP data archives. It is also one of the least common transport modes according to Eneroth et al.'s study, highly variable in occurrence over the last ten years, but most frequent during the summer months; June to September. Much of the west coast of Spitsbergen can therefore be expected to experience orographically-enhanced deposition of nitrogen from European air masses under these conditions. An example can be found in Krawczk et al. (2008), who reported a high magnitude (23 mm in 1 day), acidic (pH 4.65, representing the summer minimum) and high NO_3^- (24 μeq L^{-1}) event further south in west Spitsbergen during 2002. This event also corresponded to Eneroth et al.'s (2003) Cluster 7 owing to its association with a high pressure system over Scandinavia and a low pressure system over Iceland.

Biogeochemical response of the Midtre Lovénbreen catchment

As with any seasonally snow-covered ecosystem, the flowpaths that convey melt and rainfall through the stream network are poorly developed at the start of the summer. Therefore the timing of the June 1999 event is important because soil thermal regime and snow cover conditions will have reduced infiltration into the active layer. However, since much of the study site is dominated by glacier ice cover, delayed flowpaths through sedimentary environments at the glacier bed and its margins require consideration, as these remain unfrozen all year beneath the thicker parts of the glacier. Specifically, there are a series of crevasses at high elevation within the catchment (ca. 350–400 m: Fig. 1) which allow water to enter the glacier and be stored within sediments that remain unfrozen all year. Release from this storage is extremely slow until the annual subglacial outburst flood takes place. After this, dye tracing of the flows into the crevasses indicates residence times within the glacier that are of the order of several hours (Irvine-Fynn et al. 2005). Therefore, since liquid precipitation was observed all over the catchment, the occurrence of the June 1999 event before the subglacial outburst event makes it likely that a significant proportion of the rainfall delivered to the upper catchment entered the glacier and remained there until the outburst flood. Our data clearly show that this was the case because the first samples from MLSG were characterised by high NO_3^- concentrations. The delay between the rainfall event and the subglacial outburst (ca. 10 days) means that the nutrient recharge of subglacial sediments differed significantly from that known to occur under more "normal" circumstances, when snowmelt slowly percolates into subglacial flowpaths with lower concentrations of nitrogen (see Hodson et al. 2005b).

Figure 5c and d suggest that rapid NH_4^+ assimilation almost certainly took place during the June 1999 event because the increase in stream NH_4^+ concentrations was over far sooner than the NO_3^- increase. Further, the ratio NH_4^+/NO_3^- was almost unity in the precipitation data (1.04), but only between 0.67 (at MLW) and 0.75 (at MLE) in the streams. Downstream, at MLN, the NH_4^+/NO_3^- ratio on Day 179 was 0.69, whilst the ratio for a supraglacial stream sampled at the same time, was lower than all other sites (0.50: data not shown). Thus assimilation along active supraglacial and ice marginal flowpaths began to reduce NH_4^+ concentrations almost immediately after deposition. Given that the retreat of the seasonal snow cover across the glacier had not yet occurred, cryoconite holes were an unlikely cause of the assimilation until the snow line had retreated far beyond the ice margin: a process that began in early July (DOY 190). Wet snow, slush and near-surface talus and moraine sediments were therefore likely to have been the habitats where microbial activity was responsible for the NH_4^+ assimilation apparent in these flowpaths (Hodson et al. 2008).

The emergence of the subglacial stream during the outburst on Day 189 began as a small artesian fountain at the immediate glacier margin. Concentrations of NH_4^+ were surprisingly low given the high concentrations of NO_3^- (which indicated the presence of nitrogen supplied by the June event: see Fig. 4). The average NH_4^+/NO_3^- ratio for all subglacial waters sampled at MLSG was 0.24, and thus far lower than those described above. It is therefore evident that NH_4^+ assimilation was greater in the delayed flowpaths at the glacier bed than in the ice marginal and supraglacial flowpaths described above. Geophysical investigations of the subglacial sediments beneath Midtre Lovénbreen appear to

show the presence of coarse angular talus deposits extending along the flanks of the valley and underneath the ice (King et al. 2008). It is therefore possible that subglacial microbial communities (e.g. Skidmore et al. 2000) are very similar to those within ice marginal talus sediments because their habitats are produced from the same debris source according to King et al.'s study.

Insights from nitrogen mass balance

The different precipitation gradients ΔPpt^1, ΔPpt^2 and ΔPpt^3 produced rather different nitrogen mass balance results (Table 2). ΔPpt^1 indicated no significant $^{event}NO_3^-$ deficit (just 1.58% of the summer NO_3^- deposition and thus less than the likely errors: see Hodson et al. 2005a), whilst a marked $^{event}NH_4^+$ deficit accounted for ca. 31% of the summer NH_4^+ deposition. In contrast, ΔPpt^3 indicated deficits in both $^{event}NH_4^+$ and $^{event}NO_3^-$ (equivalent to 77 and 67% of inputs respectively), with a stoichiometry approaching 1:1. The ΔPpt^2 precipitation gradient produced an intermediate scenario to the above cases, with a deficit ratio of $^{event}NH_4^+/^{event}NO_3^-$ that was 1.5. The results for ΔPpt^1 (hereafter "Scenario 1") are therefore strongly diagnostic of a catchment biogeochemical response dominated by just NH_4^+ assimilation, whilst those for ΔPpt^3 ("Scenario 3") are diagnostic of both NO_3^- and NH_4^+ loss, perhaps by a combination of assimilation and denitrification (Hodson et al. 2005a; Wynn et al. 2006, 2007). Since it is extremely unlikely that ΔPpt^1 represented the true distribution of rainfall across the catchment, the use of either ΔPpt^2 or ΔPpt^3 (Scenarios 2 and 3 respectively) is the most hydrologically plausible. Importantly, both differ from nitrogen dynamics that have been observed at the site during subsequent years (2000, 2002 and 2003) by showing no net NO_3^- production (a process thought to be caused by nitrification and discussed by Wynn et al. (2007) at this site). Therefore, we argue that the prolonged storage of precipitation enriched with NO_3^- most likely promoted denitrification in addition to simple NO_3^- assimilation, because the former process is known to occur beneath the glacier during winter (Wynn et al. 2006). This finding, and also the fact that $^{event}NH_4^+$ assimilation was greatest along subglacial flowpaths, therefore demonstrates that subglacial ecosystems may be highly responsive to atmospheric nutrient inputs. Our study suggests that this is especially the case where crevassing allows the ingress of water from the glacier surface into a poorly developed subglacial drainage system during the early summer.

Conclusions

Meteorological analysis, three-dimensional trajectories and a 3D transport model, have been used to explore the transport patterns that gave rise to an extreme nitric acid and ammonium deposition event in Western Svalbard. The meteorology and trajectories show an airflow origin from the western part of Europe for this episode. Critically, the transport of polluted air over a stratified boundary layer minimised nitrogen loss prior to reaching Svalbard, where orographic effects caused precipitation. Similar events have since been documented elsewhere during the summer of 2002 in another part of Svalbard (Krawczyk et al. 2008) and appear likely to reoccur during any summer.

The distinct chemistry of the precipitation event during 1999 enabled us to track the fate of NH_4^+ and NO_3^- along various flowpaths within a glacier basin. Rapid assimilation occurred everywhere during the rainfall, including the glacier surface and its margins but especially along high rock–water contact environments at the glacier bed. Here sediments that are typically anoxic during the winter and early spring as a result of microbial processes were able to store a proportion of the precipitation event for up to 10 days. The storage appears to have greatly increased microbial assimilation and denitrification, yet large runoff yields still occurred during the summer. Thus the microbial foodwebs of glacial ecosystems are sensitive to atmospheric nitrogen deposition at times when terrestrial soil ecosystems can remain frozen and unresponsive. In this context, our study highlights the ecological importance, particularly during the summer melt-season, of episodic transport and deposition of reactive nitrogen in the Arctic.

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