Solute in High Arctic glacier snow cover and its impact on runoff chemistry

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ABSTRACT. The chemical composition of snow and meltwater in the 13 km^2 catchment of Scott Turnerbreen, Svalbard, was investigated during the spring and summer of 1993. This paper assesses the provenance of solute in the snowpack and its impact on runoff chemistry. Dry snow contains 420 μeq l^{-1} of solute, is slightly acidic (pH 5.4) and is dominated by Na$^+$ and Cl$^-$. Wet snow is more dilute (total concentration 340 μeq l^{-1}), and less acidic (pH 5.9). This is consistent with the elution of ions from the snowpack by meltwater. Snowpack solute can be partitioned into the following fractions: sea-salt aerosol, acid aerosol and crustal. About 98% of snowpack solute is sea salt, yielding 22 000 kg km^{-2} a^{-1}. The behaviour of snowpack-derived Cl$^-$ in runoff is distinctive, peaking at over 800 μeq l^{-1} early in the melt season as runoff picks up, before declining quasi-exponentially. This represents the discharge of snowmelt concentrated by elution within the snowpack, which subsequently becomes relatively dilute. A solute yield of 140 kg km^{-2} a^{-1} can be attributed to anthropogenically generated acid aerosols, representing long-range atmospheric transport of pollutants, a potential contributor to Arctic runoff acidification.

INTRODUCTION

Glacier snowpacks can be regarded as solute stores. Winter precipitation scavenges sea salt and acid aerosols from the atmosphere, incorporating them into the snowpack along with aeolian dust. This solute is subsequently released to catchment runoff as the snowpack melts. The purpose of this paper is to describe the chemical composition of the snowpack covering a High Arctic glacier, the provenance of the solute, and its impact on the composition of runoff from the glacier.

SITE AND METHODS

Scott Turnerbreen is a 3.3 km^2 glacier, of altitude range 230–680 m a.s.l., located in central Spitsbergen (78° 06' N, 15° 57' E, Figure 1a), occupying a catchment of total area 12.8 km^2 (Fig. 1b). The catchment geology is dominated by gently dipping Tertiary sandstones, shales and siltstones (Major and Nagy, 1972). The glacier is non-temperate throughout, and drainage is essentially subaerial (Hodgkins and others, in press a).

Sampling programme and procedure

Ninety "dry" snow samples were obtained from six pits located along the glacier centre line, at altitudes from 240 to 560 m a.s.l., between 27 April and 12 May, 1993, before seasonal melting began. The depth of the pits varied from 1.48 to 1.97 m, and a layer of depth hoar 0.19–0.28 m thick was present at the base of each. The mean density of the snowpack was 332 kg m^{-3}. Forty-one "wet" snow samples were obtained from ten pits situated near the transient snowline on

the glacier surface, at altitudes from 235 to 310 m a.s.l., between 1 June and 9 July, following the onset of seasonal melting. The depth of the pits varied from 1.58 to 0.16 m. The snow consisted largely of granular crystals 1–2 mm in dia-
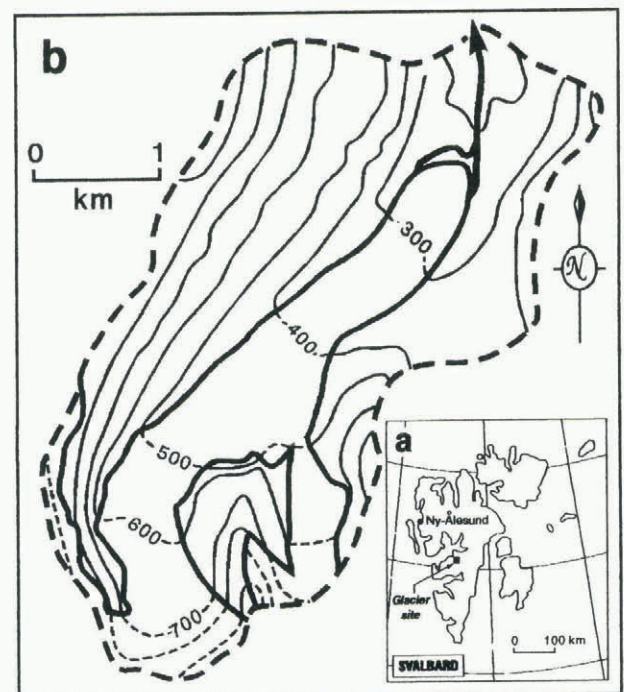

Fig. 1. a. Location of Scott Turnerbreen within the Svalbard archipelago. b. The Scott Turnerbreen catchment. Contours are in m a.s.l. Solid bold lines at the glacier snout indicate principal runoff routes.

meter. Depth hoar was absent and ice layers were frequently encountered, particularly after 18 June. The mean density of the melting snowpack during the period 1–18 June was $425 \, \text{kg m}^{-3}$. In addition, seventy-two meltwater samples were collected from the trunk stream draining the glacier, between 14 June and 10 July. The sample series commences with the onset of proglacial surface runoff. At least two samples per day were collected, at times as close as possible to minimum and maximum discharge (respectively 10.00 hours and 17.00 hours, on average).

Snow samples were sealed in polythene bags and subsequently melted by immersion in warm water, then immediately vacuum filtered through $0.45 \, \mu\text{m}$ cellulose nitrate membranes. Two 60 ml aliquots of filtrate (one for cation and Si, the other for anion and alkalinity analyses) were stored air-free in polypropylene bottles in cool, dark conditions until return to the U.K. Care was taken to pre-rinse the filtration apparatus and storage bottles with sample and filtrate as appropriate, to avoid contamination. Meltwater samples were collected in a pre-rinsed 500 ml polypropylene bottle, then immediately filtered and stored in the same way as the solid samples.

Stream stage was measured every 5 minutes with a Druck PDCR 830 pressure transducer in a stable stretch of unbraided proglacial stream, c. 250 m downstream of the confluence of the ice-marginal streams which drain the glacier. Stream stage was logged as hourly means over the period 15 June–11 July. Discrete discharge measurements were undertaken at the same location by the relative dilution method and used to obtain a rating relationship for converting stage to discharge. The typical error of the relationship is $\pm 8\%$.

Laboratory analyses

pH was determined on samples no later than 14 days after collection; air-free storage should have prevented significant change in sample pH. pH was determined with an Orion 290a portable pH meter with Ross combination electrode, calibrated with Orion low ionic strength buffers of pH 4.0 and 7.0. The temperature compensation applied was that appropriate for the measured temperature of the buffer. The precision of the measurements is ± 0.2 pH units.

The major ion and Si concentrations of samples returned to the U.K. were determined as follows. The concentrations of major base cations (Na^+, K^+, Mg^{2+}, Ca^{2+}) and acid anions (Cl^-, NO_3^-, SO_4^{2-}) were determined by ion chromatography on a Dionex 4000i ion chromatograph with, respectively, CG12/CS12 and Fast Anion-1 columns, and MSA (methane sulphonic acid) and $NaHCO_3/Na_sCO_3$ eluents. Sample loop volumes were either 50 or $200 \, \mu\text{l}$, depending on concentration. The detection limits for these determinations are approximately $0.1 \, \mu\text{eq} \, l^{-1}$. The precision of the analysis varies with the concentration of the sample, from $\pm 3\%$ at concentrations in excess of $50 \, \mu\text{eq} \, l^{-1}$ to $\pm 100\%$ at $1 \, \mu\text{eq} \, l^{-1}$.

The concentration of dissolved Si was determined by flow-injection analysis, using a Tecator FIAstar 5010 system with FIAstar 5023 spectrophotometer, V100 injector, Type III Chemifold and automatic sampler. 1 ml samples were injected into a stream of ammonium molybdate, oxalic acid and acidified stannous chloride to determine the concentration of molybdate-reactive Si, detected colorimetrically at a wavelength of 695 nm. The detection limit of this system is $1 \, \mu\text{mol} \, l^{-1}$, and the precision of the analysis $\pm 2 \, \mu\text{mol} \, l^{-1}$. Alkalinity ($HCO_3^-$) was determined by titrating 25 ml of filtered sample to an end point pH of 4.5, using 0.01 M HCl (standardized with $0.01 \, \text{M} \, Na_sCO_3$), detected with BDH mixed indicator. Titre volumes ranged from 0.10 to 6.35 ml. The precision of the analysis varies with titre volume, from ± 1 to 50 %. HCO_3^- is overestimated at low concentrations.

RESULTS

Snow composition

The composition of snow samples is summarised in Table 1. Dry snow is generally dilute, although there is considerable variability between samples, with a range of c. $5000 \, \mu\text{eq} \, l^{-1}$. Maximum concentrations correspond with a layer discoloured by aeolian dust, which was present in all snow pits. The dry snowpack is slightly acidic overall, with a mean pH of 5.4, and its composition is dominated by Na^+ and Cl^-. Si and NO_3^- were present in trace quantities, the former probably being derived from aeolian dust and the latter from acid-nitrate aerosol. Dry-snowpack solute profiles are

Table 1. Summary statistics for the compositions of dry (standard text) and wet (bold text) snow samples. Ratios of the concentrations of ions to Cl^- in seawater are from Holland (1978)

|	pH	Si $\mu\text{mol} \, l^{-1}$	Na^+ $\mu\text{eq} \, l^{-1}$	K^+ $\mu\text{eq} \, l^{-1}$	Mg^{2+} $\mu\text{eq} \, l^{-1}$	Ca^{2+} $\mu\text{eq} \, l^{-1}$	Cl^- $\mu\text{eq} \, l^{-1}$	NO_3^- $\mu\text{eq} \, l^{-1}$	SO_4^{2-} $\mu\text{eq} \, l^{-1}$
min.	5.0	0.0	4	0	0	0	0	0	0
	5.2	**0.0**	**2**	**0**	**0**	**0**	**1**	**0**	**0**
max.	6.7	1.5	2000	96	200	46	2400	7	240
	6.6	**1.5**	**1300**	**45**	**150**	**110**	**1700**	**5**	**200**
mean	5.4	0.0	170	5	19	4	200	1	25
	5.9	**0.8**	**120**	**4**	**17**	**11**	**150**	**1**	**34**
s.d.	0.4	0.0	320	12	35	9	390	1	38
	0.4	**0.0**	**230**	**7**	**33**	**24**	**310**	**1**	**54**
n	90	90	90	90	90	90	90	90	90
	29	**41**	**41**	**41**	**41**	**41**	**41**	**41**	**41**
ratio to Cl^-			0.85	0.025	0.095	0.020	1.0	0.0050	0.13
			0.85	**0.027**	**0.12**	**0.25**	**1.0**	**0.0050**	**0.23**
ratio to Cl^- in seawater			0.86	0.018	0.19	0.038	1.0	–	0.10
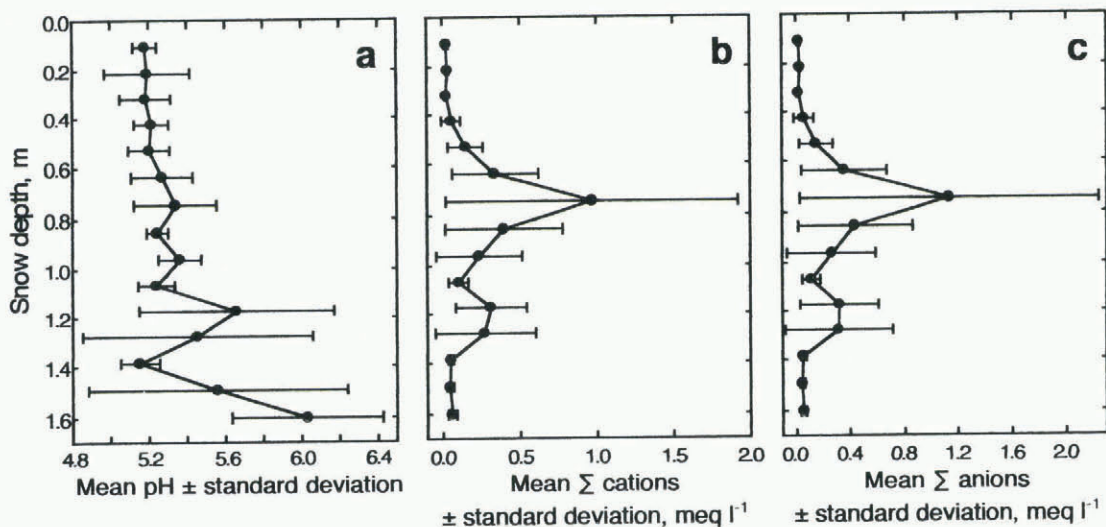
Fig. 2. Dry snow profiles. a, pH; b, cations' sum; c, anions' sum. In each case, the plotted value is the mean of six measurements in different snow pits at equivalent fractional depths; each profile has been standardized to the mean depth of the dry snowpack.

presented in Figure 2a–c; these are derived from the mean concentrations of all samples at equivalent proportional snow-pit depths ($n = 6$ for each of 15 depth intervals), standardized to the mean snow depth. Wet snow is more dilute than dry snow (mean total solute concentration 340 as opposed to 420 μeq l^{-1}), and less acidic (mean pH of 5.9). This is consistent with the elution of ions from the snowpack by meltwater (Johannessen and Henriksen, 1978; Davies and others, 1982; Brimblecombe and others, 1985; Tranter, 1991; Goto-Azuma and others, 1993b), as previously observed on Svalbard (Goto-Azuma and others, 1993a; Hodson, 1994).

The statistical significance of the differences in concentration between sample types cannot satisfactorily be tested (Hodgkins and others, in press b). A non-parametric test is required because of the highly skewed frequency distributions. A Mann–Whitney U test is not sufficiently sensitive to variations in concentration (differences of 1 and 1000 μeq l^{-1} being assigned equal importance through ranking), while a dilute sample in which all ion species are represented is not sensitively distinguished from a concentrated

sample from which one or two are absent. Furthermore, concentrations of ion species cannot be converted into mutually exclusive comparative frequency categories with sufficient cases to perform a valid χ^2 test.

Runoff composition

Significant temporal variations occur in the chemical composition of runoff. The overall trend is for the runoff to become more dilute as the melt season progresses but there are also significant variations in the relative proportions of ion species (Fig. 3a). When runoff commences on 14 June, the composition of the bulk meltwater is dominated by Na$^+$ and HCO$_3^-$, with minor proportions of SO$_4^{2-}$, Ca^{2+}, Mg^{2+} and Cl$^-$. By 23 June, the dominant ions are Na$^+$ and Cl$^-$, with minor proportions of Mg^{2+}, SO$_4^{2-}$, Ca^{2+} and HCO$_3^-$. From around 30 June to the end of the monitored period, HCO$_3^-$ again becomes the dominant anion. K$^+$ and especially NO$_3^-$ are present in only trace quantities throughout.
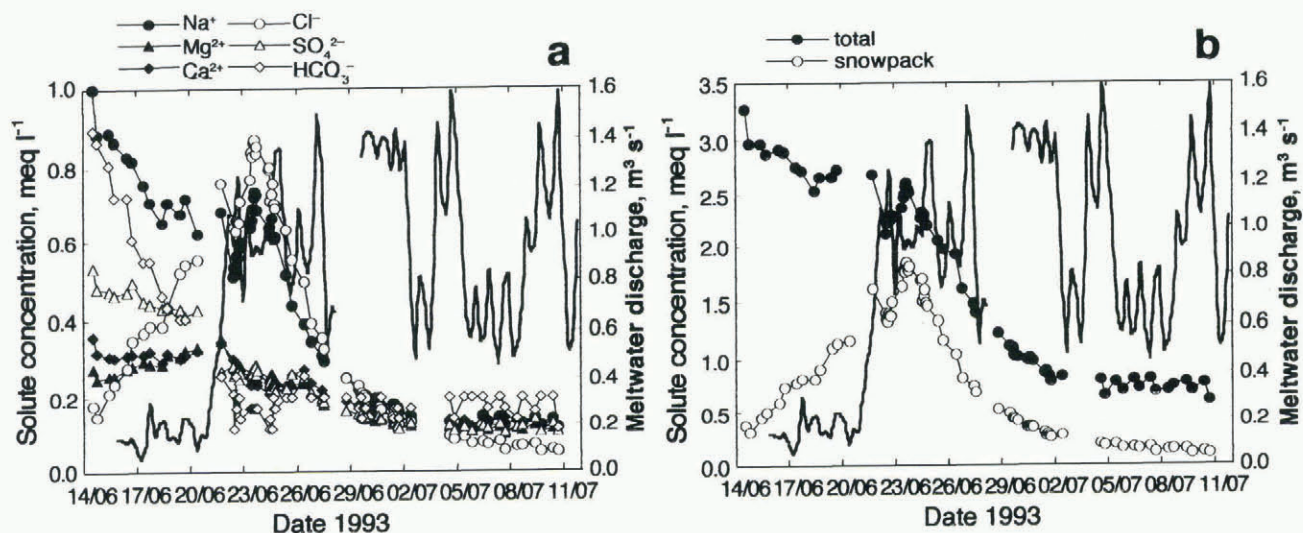

Fig. 3. Runoff time series. a. Individual ion-species concentrations and meltwater discharge; b. Total solute concentration, snow-pack-derived solute concentration and discharge.

SOLUTE PROVENANCE

Solute within the snowpack has been partitioned into sea-salt aerosol, acid aerosol and crustal components following Sharp and others (1995). The mean ionic concentrations of dry snow were the basis of the calculations, full details of which have been given by Hodgkins and others (in press b). Briefly, all NO_3^- was assumed to be aerosol-derived (precipitation-scavenged acid-nitrate aerosol) and all Cl^- to be sea-salt-derived (precipitation-scavenged sea-salt aerosol). Sea-salt contributions to Na^+, K^+, Mg^{2+} and Ca^{2+} were then determined from the Cl^- concentration using standard sea-water ratios (Holland, 1978). Residual concentrations of these ions were attributed to a crustal source (aeolian dust); Si is entirely crustally derived. A snowpack-derived SO_4^{2-} concentration was determined from the Cl^- concentration using the mean ratio (assumed constant) of these species in the pre-melt season snowpack (0.125); the sea-salt component of this concentration was determined from the standard sea-water ratio, and the remainder is apportioned to precipitation-scavenged acid-sulphate aerosol. The resulting partitioned concentrations in units of $\mu eq\, l^{-1}$ were converted to units of $kg\, m^{-3}$, and then to masses in kg by multiplying by the water-equivalent volume of the glacier snowpack. Although snow-density data are not available for all samples, mean concentrations are here believed to be a reasonable approximation to volume-weighted means, as measured snowpack density profiles are relatively uniform (standard deviation $0.05\, kg\, m^{-3}$). The glacier area is $3.3 \times 10^6\, m^2$, the mean glacier snow depth is 1.6 m (162 point measurements) and the snow density $330\, kg\, m^{-3}$ (40 measurements in four separate snow pits). The glacier snow volume is therefore $5.4 \times 10^6\, m^3$ and the water-equivalent glacier snow volume $1.8 \times 10^6\, m^3$. The results are given in Table 2. The snowpack composition is clearly dominated by sea salt, which accounts for about 98% of total solute by mass. This result is supported by the ratios of the concentrations of ions to Cl^- in the dry snowpack, which are very close to those in sea water (Table 1). The total solute mass of the glacier snowpack corresponds to a yield of $23\,000\, kg\, km^{-2}\, a^{-1}$.

SNOWPACK SOLUTE IN RUNOFF

Hodgkins and others (in press a) have described the hydrochemistry of runoff from Scott Turnerbreen in detail and have indicated that high but rapidly declining solute concentrations in the earliest days of the melt season (e.g. Na^+, HCO_3^- and SO_4^{2-}, Fig. 3a) can be attributed to solute acquisition from a proglacial icing, which represents concentrated, stored winter runoff. The behaviour of Cl^- in runoff is distinctive: its concentration increases from less than 200 $\mu eq\, l^{-1}$ when runoff commences on 14 June, to over 800 $\mu eq\, l^{-1}$ on 23 June, declining thereafter to about 50 $\mu eq\, l^{-1}$ at the end of the monitored period (Fig. 3a). Mg^{2+} follows a similar pattern, and so, to a lesser extent, do Na^+ and Ca^{2+} (Fig. 3a).

The only source of Cl^- to the bulk meltwater is the snowpack (precipitation-scavenged sea-salt aerosol). Having established that elution of solutes occurs within the snowpack, the timing of the Cl^- peak relative to the onset of sustained high discharge of meltwater strongly suggests that this represents the discharge of concentrated snowmelt. The three–five-fold concentration of Cl^- in the runoff, relative to the snowpack, is consistent with field and laboratory studies of the composition of snowmelt and its concentration

Table 2. The provenance and yield of solutes from the 1992–93 Scott Turnerbreen snowpack. Trace quantities of Si are present

Species	Sea salt	Aerosol	Crustal	Total
Na^+ (kg)	6960	0	0	6960
K^+ (kg)	251	0	97	348
Mg^{2+} (kg)	411	0	0	411
Ca^{2+} (kg)	143	0	0	143
Cl^- (kg)	12 627	0	0	12 627
NO_3^- (kg)	0	110	0	110
SO_4^{2-} (kg)	1779	359	0	2138
Si (kg)	0	0	0	0
Total (kg)	22 171	469	97	22 737
Total (%)	97.5	2.1	0.4	100
Yield ($kg\, km^{-2}\, a^{-1}$)	6728	142	29	6899

by elution within snow (Johannessen and Henriksen, 1978; Colbeck, 1981; Davies and others, 1982; Brimblecombe and others, 1987; Tranter, 1991).

This concentration peak is the product of processes that concentrate solutes on the surface of individual ice grains, i.e. melt–freeze cycles (Bales and others, 1989) and vapour transfer (Cragin and others, 1993), and the initial distribution (Bales and others, 1989) and subsequent redistribution by percolating meltwater (Colbeck, 1981) of solutes within the snowpack. Snowmelt therefore becomes relatively dilute for the remainder of the melt season. The snowpack-derived concentration of a given ion in the runoff, $C_{X-snow/runoff}$ can be estimated from the product of the concentration of Cl^- in runoff, $C_{Cl-snow/runoff}$, and the ratio of the ion to Cl^- in the dry snowpack, K, assumed to be constant, i.e. $C_{X-snow/runoff} = K[C_{Cl-snow/runoff}]$. The assumption that K is constant implies that significant *preferential* elution of ion species does not occur; reference to the ratios of ion concentrations to Cl^- in Table 1 suggests that this is reasonable. Total and snowpack-derived solute concentration time series are shown in Figure 3b, which illustrates the rapid early melt-season "flush" of concentrated snowpack solute in runoff, and its subsequent dilution.

DISCUSSION

The results presented in this paper confirm the dominance of sea-salt-derived solute in the snow cover of Svalbard. Arkhipov and others (1992) sampled the snow cover on Kongsvegen and Amundsenisen, and concluded that sea-salt aerosols dominated the composition of the snowpack. Semb and others (1984) concluded that the observed concentrations of ions in snow were consistent with precipitation scavenging from a near-surface atmospheric layer, and that sea salt was the dominant source of solute. By partitioning snowpack solute according to provenance, this paper has quantified the relative contributions of sea-salt aerosol, acid aerosol and aeolian dust to the snowpack: 97.5, 2.1 and 0.4%, respectively (Table 2).

The composition of snow sampled at Scott Turnerbreen prior to and following the onset of seasonal melting is consistent with the elution of ions from the snowpack by meltwater. The results presented in this paper enable the impact

of this process to be traced quantitatively from the snow-pack to the runoff. The proportion of snowpack-derived solute in total meltwater solute is 12% when surface runoff commences on 14 June; it increases linearly to a maximum of 75% on 24 June, then decreases quasi-exponentially to 17% at the end of the monitored period on 10 July (Fig. 3b). The contribution of snowpack solute to runoff can therefore be demonstrated to be a brief but intense concentration episode early in the melt season.

Goto-Azuma and others (1993b) found that 10–60% of snowpack SO_4^{2-} at Austre Brøggerbreen in Svalbard was from non-sea-salt sources. Other measured non-sea-salt SO_4^{2-} concentrations in snow from Svalbard are in the range 4.5–11 μeq l^{-1} (Gjessing, 1977; Semb and others, 1984). The results presented in this paper quantify the amounts of solute derived from sources other than sea salt, principally acid aerosols. The snowpack acid aerosol solute yield amounts to 140 kg km^{-2} a^{-1}, or 2.1% of the total snowpack solute yield (Table 2). The High Arctic islands are sinks for long-range, trans-boundary atmospheric pollution (Hansen and others, 1996). The supply of atmospheric pollutants to Svalbard exhibits strong seasonality, and is at a maximum during February–April (Rahn and McCaffrey, 1980; Joranger and Ottar, 1984), corresponding to the winter precipitation maximum in this part of the archipelago (Hanssen-Bauer and others, 1990). Winter prevailing winds in the Svalbard archipelago are easterly and southeasterly (20% and 40% frequencies, respectively; Hanssen-Bauer and others, 1990), i.e. from the direction of industrial Eurasian regions. Deposition of precipitation-scavenged acid sulphate and nitrate aerosols therefore occurs in the Svalbard snowpack and is a potential contributor to meltwater acidification. A three–five-fold increase in H$^+$ in runoff, consistent with the concentration of solutes due to snowpack flushing described in this paper, could decrease the mean pH of runoff from 7.3 to 6.6–6.8, other factors remaining equal (since -log$_{10}$(H$^+$) = pH, where H$^+$ is in units of mol l^{-1}). Though aquatic ecosystems are not typically well developed in glacierized catchments, the buffering capacity of such catchments is also characteristically limited, so meltwater acidity is unlikely to be effectively neutralized.

ACKNOWLEDGEMENTS

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