THE COMPARATIVE FLUXES OF STRONG-ACID ANIONS IN MELTING SNOWPACKS AND SURFACE WATERS DURING SPRING MELT IN A BOREAL FOREST

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ABSTRACT

The chemical evolution of meltwater from the snowpack, intermittent streamwaters and subsurface waters from the Lac Laflamme watershed (Québec) during the spring melt indicates that the upper organic podzolic soil layer (A_0, A_1) is an important factor in the control of NO_3 concentrations in surface waters. On the other hand, SO_4^{2-} concentrations in the streams are also dependant on the chemical characteristics of water in the lower soil layers (B_1, B_2) although the upper organic layer does seem to exert a buffering effect on the concentration of this ion. The soil horizons also act as an additional source for SO_4^{2-} over and above the SO_4^{2-} loads deposited by rain and discharged by the pack during the melt period. The total load of SO_4^{2-} discharged to the ground during the melt period was 25 ± 8 meq m^{-2} while the total load exported by the surface water was 43 ± 36 meq m^{-2}. In the case of NO_3, however, the nature of the role that the soil may play either as a source or sink of this ion is not clear. The ground received a discharge of 10 ± 7 meq m^{-2} of this ion during the spring melt while the exported load from the soil to the surface waters was 10.8 ± 9 meq m^{-2}.

INTRODUCTION

Many studies have shown that the acidity of surface waters increases significantly in watersheds sensitive to acid precipitation during the snow melt period (Seip et al., 1980; Johannessen et al., 1977, Marmorek et al., 1984). The same phenomenon has been shown to occur regularly in the Lake Laflamme watershed situated in the Laurentian Park north of Québec city (Charette et al., 1983). The acid pulse which passes through the lake basin under the ice-cover during the melting of the snowpack originates from two main sources. The first is the early direct runoff from the melting snowpack on the shore of the lake (Jones et al., 1984) in which preferential leaching of the snow by the meltwater leads to high concentrations of acidic polluants in the discharge at ground level. The second source is the surface runoff generated by interaction between meltwater and soil horizons which finally enter the lake via the main tributary and numerous intermittent streams; the latter appear when the water table rises, in such events the infiltration of meltwater into the soil is shallow and restricted principally to the upper soil horizons (Jones et al., 1985).

The acidity of the meltwaters discharged from the pack is due primarily to the presence of the strong-acid anions SO_4^{2-} and NO_3. In the subsequent contact between the soil and the meltwaters different geochemical processes such as ion-exchange, adsorption and desorption, and leaching can cause both partial neutralization of the soil solutions and modify their overall chemical composition (Wiklander, 1980; Cronan, 1980; Wiklander, 1975). In

addition, physical precipitation from solution and in particular, micro and macrobiological uptake can temporarily remove certain ions such as NO_3 from solution although SO_4^{2-} seem to be more persistant in solution (Jones et al., 1984). Thus SO_2^{2-} is usually the major strong acid anion in surface runoff and generally acknowledged to be the main cause of acidity in these waters.

The acidity of runoff, however, is not only a function of the ionic load per unit area originally deposited on any particular basin but rather the result of the high ionic fluxes (load area^{-1} time^{-1}) which are produced at certain critical periods of the hydrological cycle (ex: spring melt, autumnal rains). Papineau (1985) has shown in effect, that during May 1984 when snow melt was maximal at Lac Laflamme 34% of the annual flow for that year passed through the lake transporting in the process 36% of the SO_4^{2-} annual load exported by the lake (32 kg ha^{-1} = 64 meq m^{-2}) and 48% of the NO_3 annual load (3.76 kg ha^{-1} = 6.06 meq m^{-2}). However, 60% of the H+ annual load (0.0087 kg ha^{-1} = 0.87 meq m^{-2}) was transported from the watershed at this time.

As a first step in the study of the acid pulse in lake Laflamme during snowmelt we wished to determine the contribution of the total strong-acid anion load which had accumulated in the snowpack during the winter prior to the beginning of the spring melt, and from the rain-on-snow events during the melt period, to the strong-acid anion flux in the surface runoff. The difference between the measured loads in the snowpack (plus precipitation) and in the surface waters is then used to determine the extent to which the soil - subsurface water system acts as a sink or as a reservoir of strong-acid anions during the spring melt period.

Site description

The watershed of Lake Laflamme (0.684 km^2, lake basin, 0.061 km^2) is situated (46°11'N, 74°57'0) 80 km north of Québec City. The mean annual level of the lake is 777 m and the highest point in the watershed is at 884 m. The watershed which lies wholly in the Montmorency Experimental Forest is covered with conifers; balsam fir is the predominant species associated with a small amount of black spruce and white birch. The soil is a humic podzol covering a thick layer of till and moraine which in turn overlays the precambrian gneiss of the Grenville Formation.

Mean annual temperature is 0.2°C (minimum -15°C, January; maximum, 15°C July). Annual precipitation is 1 400 mm, snow 490 mm (Plamondon, 1981). The hydrographic network is a very poorly developed system with one main tributary which enters the lake via a marsh. Two secondary tributaries which usually run dry in winter are supplemented by numerous intermittent streams during the spring melt period. Azzaria et al. (1982) has shown that groundwater is an appreciable source of water for the lake during much of the year except during spring when surface runoff is important.

Sampling

All sampling was carried out in the winter of 1983-1984 and spring 1984 on the northern slope of the Lake Laflamme watershed (Figure 1). Wet precipitation was collected using wet-only automatic samplers (Sangamo). One sampler was placed in an open clearing the other under the balsam-fir canopy. The snowcover was sampled at three separate locations, an open site, a balsam-fir covered site and a partially covered balsam-fir site. Snow pits were used to take integrated samples of the snowcover for chemical analysis. The water equivalence (WE) of the snowcover was calculated from density measurements of a vertical profile of the pits using a Swedish density gauge (AB Production meter, Stokholm). The calculations were verified by comparing the calculated WE values with the volumes of melted snow from the integrated samples. Meltwaters issuing from the pack were collected directly using lysimeters (1 m^2) of fiberglass (Jones and Deblois, 1986) and funneled via plastic (ABS) tubing into covered plastic recipients (45 L) installed deeper in the soil horizons.

Surface runoff was collected as grab samples into plastic bottles at a temporary gauging station that was set up to measure the flowrate of a major intermittent stream. Subsurface water was pumped from a depth of 1 m up through a piezometer tube by means of a peristaltic pump.

Daily samples were taken between the 24 April to 24 May during the main spring melt. Sampling frequency was lower before the 24 April during the partial melt and after the 24 May when the intermittent streams started to dry up.

All samples were analysed for the strong-acid anions Cl^-, SO_4^{2-}, NO_3^- within 48 hours of sampling (conservation temperature $4°C$). The samples were filtered (0.45 μm. nucleopore) and the analyses were carried out using a Dionex 12S ion chromatography unit. (H.G. Jones et al., 1985).
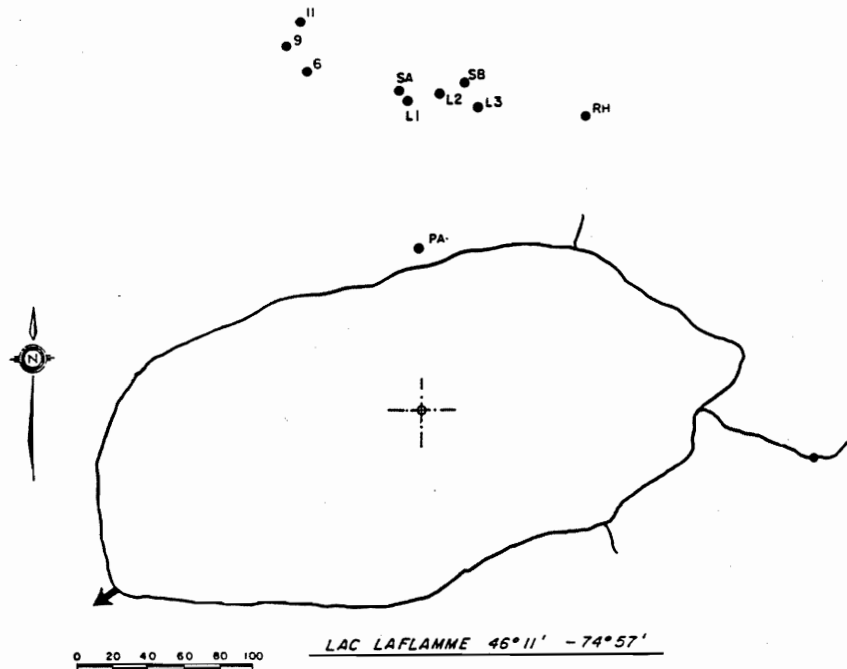

Figure 1: Study site (Lac Laflamme, Laurentian Park Québec), spring 1984: SA, SB, Wet-only collectors; L1, L2, L3, lysimeters; 6, 9, 11, snowcover sampling sites; RH, intermittent stream; PA, pieaometer.

Meteorological data

Some quantitative data on the precipitation were not recorded due to technical difficulties. Data obtained at the Lake Laflamme site was thus supplemented by data from the meteorological station at the Forêt Montmorency Research Station 1 km from the study site. Plamondon (1982) has already demonstrated the very significant correlations that exist between the meteorological data taken at the Lake Laflamme site and those recorded at the Forêt Montmorency Research Station.

Hydrological data

The hydrological data was obtained from the Lake Laflamme data bank of the Inland Waters Directorate, Environment Canada (Québec région) as compiled by J. Stein and J. Roberge of Laval University (Stein and Roberge, personal communcation and Environment Canada, Inland Waters Directorate, Québec region). The data available are the continuous record of meltwater discharge and streamwater flow rate and the fluctuations in height of the water table.

Chemical data

Qualitative data on two melt periods were not available. These were the rain-induced melts of 20-22 March and 6-7 April. The concentrations of strong-acid anions in these meltwaters were thus calculated from the data on the precipitation and those generated by the model SNOQUAL 1 (Jones et al., 1986) from the chemical and hydrological data available on the snowpack.

Results and discussion

Concentrations of Cl^-, SO_4^{2-} and NO_3^- in meltwaters, streamwaters and subsurface waters

Figures 2, 3 and 4 show the change of concentrations with time of Cl^-, SO_4^{2-} and NO_3^- in meltwaters (weighted means of the three lysimeters), surface runoff (gauged intermittent stream) and subsurface waters during the period February - May 1984. A mid-winter melt observed in February (9-16) was followed by two short melt periods, 20-22 March and 6-7 April; all these melts were provoked by warm periods with heavy rain (15 February, 39 mm; 21 March, 14 mm; 6 April, 22 mm of rain). The main melt started to discharge continously from the 11th April except for diurnal melt-freeze cycles and the occasional cold period (ex: 2-5 May).

The evolution of the concentrations of all the strong-acid anions in the meltwater shows the progressive decrease in value that is due to the leaching of pollutant aerosols. These are associated more with the surface of the snow grains than with the main body of the ice matrix (Johannessen and Henricksen, 1978). In the later stages of the melt when frequent rain-on-snow events occur, the concentrations in the discharge are more dependant on the concentration of the ions in rain than on the residual ionic concentrations in the snow grains.

In contrast to the meltwaters discharged from the pack, the chemical dynamics of the ions in surface and subsurface waters differs appreciably. Both Cl^- and SO_4^{2-} (Figures 2 & 3) concentrations decrease significantly in the subsurface waters in the melt from the 16 April to 1 May while NO_3^- concentrations rise from values which are below the detection limit of our analytical method to a maximum value of 15 $\mu eq \ L^{-1}$ (figure 4). From 1 May to the 24 May, Cl^- concentrations in the subsurface waters closely follow those in the intermittent stream decreasing regularly to a value of 5 $\mu eq \ L^{-1}$ while SO_4^{2-} values in the stream are generally 15-20 $\mu eq \ L^{-1}$ lower than those in subsurface waters which oscillate around a concentration of 130 $\mu eq \ L^{-1}$. Concentrations of NO_3^- in the streamwaters decrease in an exponential fashion during the whole melt period until they reach very low values (10 $\mu eq \ L^{-1}$) at the moment when the intermittent streams are drying up. By the end of this period, the NO_3^- concentrations in the subsurface waters have decreased back again to negligable amounts.
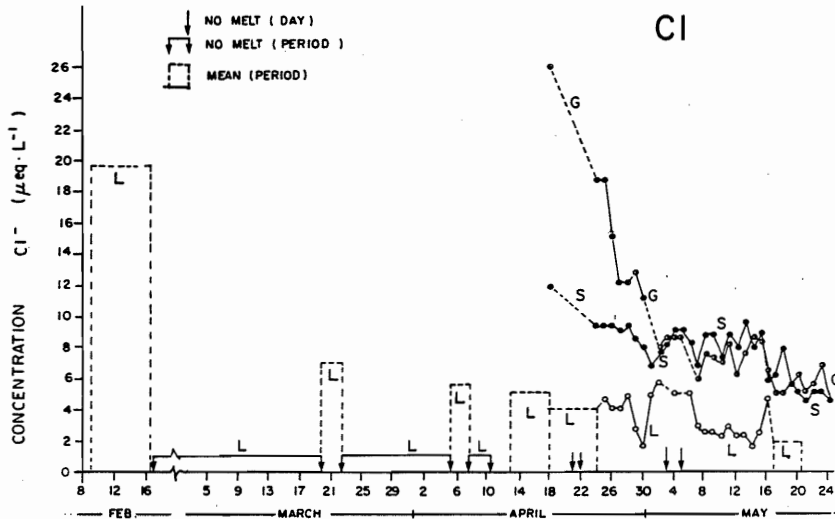

Figure 2: Concentration of Cl^- in meltwaters (L), intermittent streamwater [S] and subsurface water [G], Lac Laflamme, spring, 1984. Values for meltwaters on 22-27 March and 6-7 April are the mean of estimated concentrations (see text, chemical data).

48

Figure 3: Concentration of SO_4^{2-} in meltwaters (L), intermittent streamwater [S] and subsurface water [G], Lac Laflamme, spring, 1984. Values for meltwaters on 22-27 March and 6-7 April are the mean of estimated concentrations (see text, chemical data).
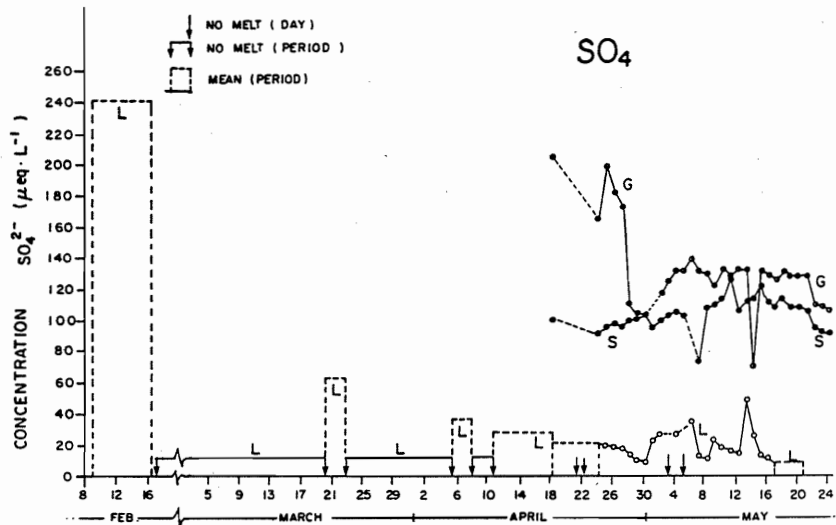

Figure 4: Concentration of NO_3^- in meltwaters (L), intermittent streamwater [S] and subsurface water [G], Lac Laflamme, spring, 1984. Values for meltwaters on 22-27 March and 6-7 April are the mean of estimated concentrations (see text, chemical data).
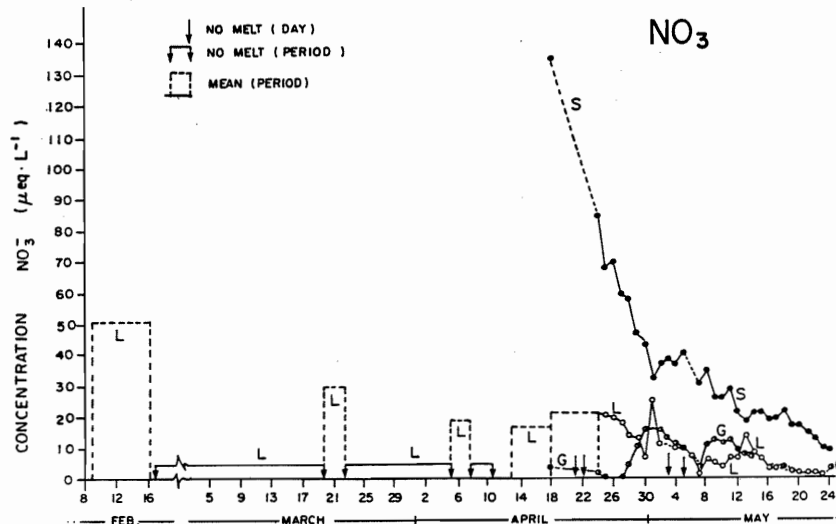
To explain the evolution of surface water chemistry we accept "a priori" the simple hypothesis that the intermittent surface runoff at Lake Laflamme is generated by waters which cannot infiltrate below the organic horizon (A_0, A_1), when the meltwater discharge rate is superior to the infiltration rate into the soil, and/or to the resurgence of subsurface water from lower soil horizons (A_2, B_1, B_2, C) up through the organic layer when the water table is high. The processes occur concomitantly; the extent to which each one contributes to stream flow being dependant on meltwater discharge rate and the height of the water table (Sklash and Farvolden, 1979). In this hypothesis we do not consider any surface runoff arising from direct runoff on frozen ground because the forest floor is rarely completely frozen and many opportunities for infiltration exist even in this type of condition (Price and Henrie, 1983). Neither do we consider the direct discharge of subsurface water from mineral soil into the streambeds due to the fact that the

intermittent streams run in depressions in the organic soil, change flow direction with flowrate, and can only be reasonably gauged in well defined gullies where flow direction becomes independant of flowrate. Finally, we consider the lower soil horizons as a homogenous matrice with no appreciable influence on the spatial distribution of subsurface resurgence toward the surface. The water table level is taken to be the only factor which determines the area of the basin where resurgent flow occurs. This latter part of the hypothesis, however, should be treated with caution as we know that pipeflow can occur in the Lake Laflamme watershed (Roberge, personal communication).

Except for a very small amount of surface runoff on the 18 April the intermittent stream began to run on the 24 April. This coincided with the first large meltwater discharge from the lysimeters on the 23-29 April when an average discharge rate of 22 mm/day was recorded. During this period, some of the meltwater ran rapidly through the organic horizon to the stream bed leaching some Cl^- in the process, while the rest penetrated to the lower soil mineral horizons to dilute the concentration of Cl^- in the water table (Fig. 2). Under the influence of increased infiltration, the water table rose and became progressively diluted in Cl^- until the water table reached, or rose very close to, the organic horizon over most of the study area of the watershed on 30 April - 1 May (Jones et al., 1985). At this point, the two pathways of surface flow to the stream become indistinguishable and, in the case of Cl^-, the concentrations of surface water and subsurface are very similar. This would suggest that resurgence of subsurface water through the organic layer has very little effect on the chemical composition of the subsurface water and the quality of the two waters becomes almost identical when the water table attained its highest levels on 30 April - 2 May, 9 - 12 May and 22 May. In the case of SO_4^{2-} (Fig. 3), both subsurface waters and streamwaters have also the same quality on the above mentioned dates. However, during the intervening periods when the level of the water table drops towards the mineral horizons, the chemical composition of the streamwater reflects the relatively greater amount of meltwaters having had only contact with the surface organic soil. These results suggest that a greater amount of SO_4^{2-} is leached from the lower soil horizons (A_2, B_1, B_2) than the organic, they also indicate that contrary to Cl^- and NO_3^-, the concentrations in the stream remain relatively constant due possibly to some mechanism of SO_4^{2-} buffering by the upper soil layer (Singh, 1984). The only major deviation of stream water quality from the overall evolution of SO_4^{2-} concentrations occurred on the 7 May when flowrate increased rapidly after a cold period. In the case of NO_3^- (Fig. 4) it is apparent that the organic layer plays a dominant role in the control of the concentrations of this ion in the streamwaters. The leaching of NO_3^- from the organic layer by the dilute meltwaters gives rise to very high concentrations in the streamwaters and also increases the NO_3^- concentrations in the subsurface water which were originally negligible. As the melt progresses both subsurface, and surface water NO_3^- concentrations drop to very low values. The behaviour of NO_3^- in the system is probably the net result of two processes. The first is the leaching of NO_3^- which has been produced in the upper layers by the microbiological degradation of organic nitrogen during the winter and early spring. This is followed by the uptake of NO_3^- by the vegetative layer and root zones both in and below the organic soil horizon as plants respond to the warm and humid conditions of the spring season.

Total fluxes of Cl^-, SO_4^{2-} and NO_3^- in meltwater discharge during the whole melt period.

Although an examination of the relative ionic concentration in figures 2, 3 and 4 shows in the case of every ion that the daily fluxes in streamwaters are greater than the fluxes discharged from the pack, this does not necessarily hold true for the average flux over the whole melt period (Feb. 8 - May 24). In the first part of the melt, the meltwaters which have very high concentrations of all ions infiltrate the soil and are retained until the streams start to flow. In order to determine if the fluxes for surface waters and meltwaters are similar for the whole period, the measured load (meq m^{-2}) discharged from the snowpack into the field lysimeters is compared to the calculated load (meq m^{-2}) exported from the drainage basin of the intermittent stream. There are, however, two problems in calculating values for the latter. The first is that the total ionic load which passed through the gauging station is calculated from the concentrations of grab-samples taken at one instant in time and at a particular flowrate. To calculate the total load discharge by the intermittent stream over a 24 hr period the average concentration of the two successive samples was used; daily loads were then summed over the whole discharge period. The second problem concerns the determination of the area (m^2) of

the drainage basin of the intermittent stream; this information is necessary to translate the ionic flux of the surface water into a form compatible with that of the meltwater flux (meq m^{-2}).

The areas of the drainage basins of the intermittent stream are highly variable, they can increase or decrease according to the intensity of precipitation, meltwater discharge and the height of the water table. They are extremely difficult to measure directly and thus have to be estimated by other means. To estimate the mean area of the drainage basin of the gauged intermittent stream for the whole melt period we use two models; the balanced Cl^- budget (hydrochemical model) and the proportional hydrological flux model. In the balanced Cl^- model we consider that Cl^- is a conservative ion that passes rapidly through the system by hydrological transport (Skartveit and Gjessing, 1979; Overrein et al., 1976). The area of the drainage basin is then calculated from the following equation.

$$A_S = \frac{[C]_S \, V_S}{L_L}$$

where A_S = mean area of the intermittent stream during the melt period, m^2;
$[C]_S$ = mean concentration (volume weighted) of Cl^- in the stream, meq, m^3;
V_S = total volume discharge of the stream, m^3;
L_L = total ionic load of Cl^- discharged by the lysimeter meq m^{-2}.

The estimated mean area using this model gave a value of $1.1 \pm 0.7 \times 10^3 m^2$ for A_S. (table 1).
The standard deviation around the mean is the result of the error in the calculation of the total Cl^- load discharged from the lysimeters during the melt periods 20-22 March and 6-7 April.

The second model assumes that the drainage basins of all tributaries are proportional to the flow rates of the tributaries at any one time. The mean area of the intermittent stream for the whole melt period can thus be estimated from equation 2.

$$A_S = \frac{V_S A_{LL}}{V_{LL}}$$

where A_{LL} = the area of the Lake Laflamme watershed, $68 \times 10^4 m^2$;
V_{LL} = 1.12 x the total volume discharged from the lake during the melt period (m^3).
The conversion factor (1.12) is necessary to correct for the ground water losses that occur in this system due to subsurface flow under the lake (Papineau, 1985).

The estimated mean area using this model gave a value of $2.03 \times 10^3 m^2$.

Table 1: Mean area (As) of drainage basin of the intermittent stream as calculated from two models, spring melt, Lake Laflamme 1984.

MODEL	A_S, ($\times 10^3$) m^2
Hydrochemical model (Cl^-)	1.10 ± 0.7
Proportional hydrological model	2.03

From the total load of SO_4^{2-} and NO_3^- (meq) discharged by the intermittent stream during the melt period and the areas estimated from the hydrochemical and hydrological models the export of SO_4^{2-} and NO_3^- (meq m^{-2}) can be calculated.
The total flux (meq m^{-2}) of SO_4^{2-} and NO_3^- for meltwaters and streamwaters during the whole period are recorded in table 2. Although the differences between the meltwaters and surface waters are not statistically significant (t, test) due to the large standard deviations around the mean values, the results do indicate that the soil-subsurface water system is more probably a source of SO_4^{2-} (approx. 10-20 meq m^{-2}) than a sink during the spring melt. As it is unlikely that till and sand retain SO_4^{2-} by chemical processes and the probable source of the additional SO_4^{2-} in the surface waters is the B_1 and B_2 soil horizons; these horizons contain large quantities of humic matter and Fe respectively. In the case of NO_3^- the calculated loads in the streamwaters (14 ± 9 meq m^{-2}, hydrochemical model; 7.6 meq m^{-2}, proportional hydrological model) are closer to the meltwater load discharged to the soil by the lysimeters (10 ± 7 meq m^{-2}) and we cannot suggest at this time as to whether the soil-subwater system is a sink or a source during the spring melt.

Papineau (1985) has calculated the mean annual SO_4^{2-} and NO_3^- loads exported from the watershed for 1981-1984. In the case of NO_3^- the mean annual load (4.47 kg ha$^-$ = 7.21 meq m^{-2}) is far inferior to the mean annual load deposited by wet precipitation (22 kg ha^{-1} = 35.5 meq m^{-2}); NO_3^- is thus stocked in the system as organic N. In contrast, the mean annual load of SO_4^{2-} exported by the lake (32 kg ha^{-1} = 64 meq m^{-2}) is much closer to the total wet deposition of 28 kg ha^{-1} (57.1 meq m^{-2}). If one takes into account the groundwater losses (12%) and the estimated dry deposition loading to the watershed (15-40% of total load) the net chemical budget of SO_4^{2-} in the watershed would indicate that some SO_4^{2-} is retained in the system. The sinks may either be the upper soil horizons and/or the lake sediments. This hypothesis is conditional to the development of methods to obtain more precise data on dry deposition in this watershed. Our results do suggest, however, that during the spring melt the high hydrological flux through the system causes a reversal of the role that the soil generally occupies in the annual geochemical cycle of these elements at Lac Laflamme.

Table 2: Strong-acid anion budgets for meltwater (+ precipitation) and surface runoff at Lac Laflamme, spring 1984.

INPUT OUTPUT	PERIOD	LOAD meq m^{-2}		
		Cl$^-$	SO_4^{2-}	NO_3^-
INPUT BY SNOWMELT AND PRECIPITATION	Dec. 83-20/05/84	3.6 ± 2.4	25 ± 8	10 ± 7
OUTPUT CALCULATED FROM HYDRO-CHEMICAL MODEL		3.6	55 ± 36	14 ± 9
OUTPUT CALCULATED FROM HYDRO-LOGICAL MODEL		1.9	30	7.6
MEAN OUTPUT STREAMFLOW		2.8 ± 2.4	43 ± 36	10.8 ± 9

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