Nitrogen Dynamics and Sub-Ice Meltwater Patterns in a Small Boreal Lake During Snowmelt

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ABSTRACT

A study of the chemical composition of snow meltwaters, stream waters, runoff, the water column under the ice cover and the discharge of a small boreal lake was undertaken during the spring melt 1988. The results led to the characterization of the main factors for the episodic acidification of lake sub-ice waters. Increase in the acidity of the waters is due in large part to the dilution of base cation (Na, K, Ca, Mg) concentrations and of the strong-acid anions SO_4 and Cl. The behaviour, however, of the other strong-acid anion, NO_3, does not follow the same pattern. During the early melt, a strong pulse of acidity was associated with relatively high concentrations of NO_3. Conversely, a second pulse of acidity during the main melt is accompanied by waters which contain very low concentrations of NO_3. This NO_3 pattern is due to two major origins of the acidic pulses i.e snow meltwaters and surface runoff. The first pulse is due to the early input of concentrated snow meltwaters from snowbanks on the lakeshore. The second pulse is due to surface runoff in the upper soil horizons (throughflow). The removal of NO_3 from meltwaters routed to the lake through these soil horizons may be due to denitrification in the soil and assimilation in the root zone during meltwater infiltration. NO_3 removal may also occur in the sub-ice waters due to assimilation by phytoplankton.

INTRODUCTION

First studies on the effect of acid rain on the environment concentrated on sulphur emissions (SO_2) and precipitation (SO_4). In recent years, however, more attention is being paid to the emission of N-species (NO_x, NO_y) and the impact of NO_3 (Kelly et al, 1990). The study of the effects of N-species on aquatic ecosystems is complex because NO_3 is not only a strong-acid anion but also a major nutrient source for catchment areas. Transformation and assimilation of N-species within ecosystems can increase or decrease the ambient acidity depending on the form (e.g. NO_3, NH_4) and the type of biological activity.

Environment Canada has been carrying out an intensive study since 1980 on the pathways of SO_4, NO_3 and other major ions through a small boreal lake (Lac Laflamme) 80 km north of Quebec City (Papineau and Haemmerli, 1992). Emphasis has been placed on the springmelt period when up to 20-30% of the annual input of acidity, accumulated in the winter snow cover, is discharged by snow meltwaters within a short period (2-4 weeks).

During the springmelt Lac laflamme supports an ice cover (~1 m). Ice cover constitutes a physical barrier between the lake and the atmosphere which ensures the thermal stratification and vertical stability of the water column (Bergmann and Welch, 1985). Melting ice cover also contributes to the chemical

composition of sub-ice waters. Ice cover can thus play a major role in dictating flow patterns of acidity within the lake basin during the snowmelt season (Henriksen and Wright, 1977; Hultberg, 1977; Prentki et al., 1979; Jeffries et al., 1979; Hendry et al., 1980; Charette et al., 1984; Jones et al., 1984; Gunn and Keller, 1985, 1986; Booth et al., 1987; Adams and Allan, 1987; Molot et al, 1990; Roberge and Jones, 1991).

The purpose of the study described below was to investigate the inputs and sub-ice flow-through patterns of acid waters in Lac Laflamme during the springmelt. Emphasis was placed on the origins of NO_3 and its relationship to the acidity of the stratified lake waters. The field work was conducted in the spring of 1988.

METHODOLOGY

Study site
Lac Laflamme is a shallow headwater lake (6.1 ha, 780 masl) in the boreal forest 80 km north of Québec city; the area of the catchment is 68.4 ha (Figure 1). The volume of the lake is $125*10^3$ m^3 and its depth does not exceed 5.3 m. There is one main tributary which assures a continual but very low flow in winter. During the spring melt the hydrologic input is supplemented by several intermittent streams. The average annual mean temperature and precipitation are $0.2^{\circ}C$ and 1424 mm respectively. Nearly one third of the precipitation falls as snow. At the end of the accumulation season the forest snowpack typically reaches 100 to 150 cm depth and 350 to 400 mm of water equivalent. The melt season generally extends from the end of March to late May and ice-off occurs in mid-May.

Methods
A network of sub-ice access holes was installed in February and March 1988 along the northern shore of the lake. Each hole collared a white plastic tube which prevented mixing of lake waters with meltwaters on the ice cover. 25 holes were sited at different points on the lake surface. For the purposes of this particular study on NO_3 dynamics we have used the data from access hole B20

only, situated 20 m due south of a point on the lakeshore 160 m from the lake discharge (Roberge and Jones, 1991).

The sampling period covered the Julian day (JD) interval 103 (April 12) to 139 (May 18). Lake waters were sampled at the lake discharge and at access hole B20. Sub-ice water temperatures ($\pm 0.1^{\circ}C$) were measured with a digital thermocouple thermometer (Cole-Parmer) on the end of a probe; water samples were drawn by a syringe (60 cm^3) up through a tygon tube (3 mm) mounted on a similar probe (Roberge and Jones, 1991). Throughflow (meltwaters routed rapidly to the lake via the upper horizons of the soil when the water table is high) samples were taken at 3 sites on the shoreline. The frequency of sampling varied from daily to biweekly according to snow meltwater production; the ice cover was sampled intermittently.

Samples were filtered (0.2 μm); conductivity and pH were measured by the use of a Radiometer conductivity meter (CDM) and pH meter (pHM 26) respectively. The remainder of the sample was conserved for chemical analysis (cations, H_2SO_4; anions at $-10^{\circ}C$).. Anionic concentrations (SO_4, NO_3, Cl) were determined by the use of an ion chromatograph (Dionex 2010); cations (Ca, Na, Mg, K) were determined by atomic absorption. NH_4 and HCO_3 were not determined; no validation of ionic balance was therefore carried out. Some data on the composition of pre-melt lake waters were made available by Environment Canada. They represent the results of weekly sampling at the centre of the lake and at the outlet in February and March (Couture, 1991).

In two separate experiments rhodamine WT was used to trace the pathways of snow meltwaters and throughflow waters through the lake basin. 1L of 20% solution was poured into a trench (0.3 m x 1m x 0.45 m depth) in a snowbank 2 m back off the lakeshore and 165 m from the lake discharge almost due north of B20 on JD 99. On JD 29, 2 L of the same solution was poured into the throughflow gauging site 46 m back off the lakeshore at a point 300 m along the northern shore from the lake discharge (Roberge and Jones, 1991).
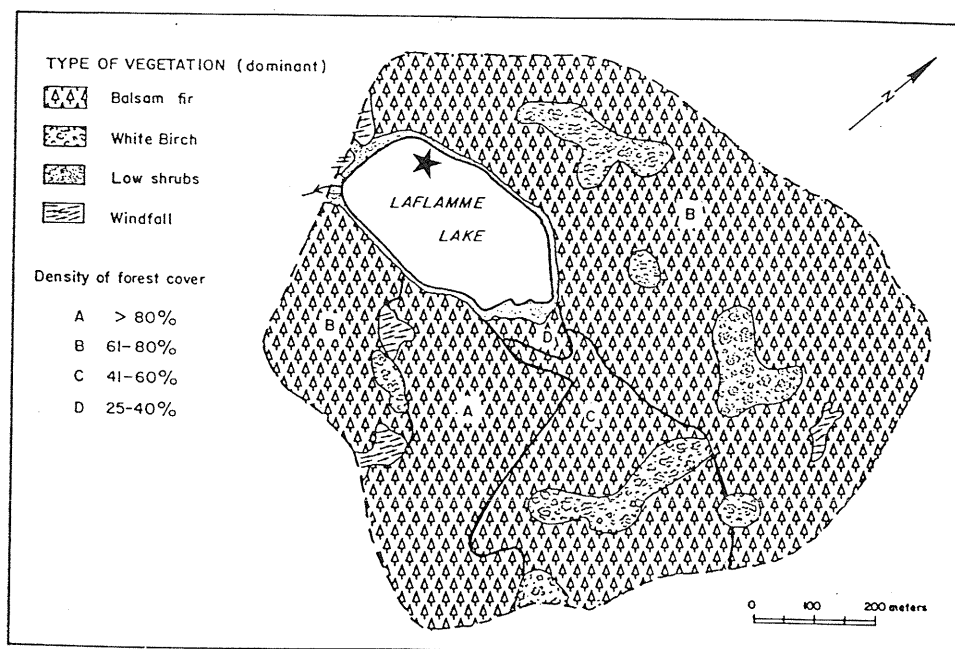
Figure 1. Lac Laflamme: vegetative cover of the catchment area, location of lake discharge and study site for the sub-ice profile measurements.

RESULTS AND DISCUSSION

Meltwater hydrology

The melt period occurred between March 26 (JD 85) and May 18 (JD 139). No appreciable amount of rain fell during the melt but a total snowfall of 61 cm was recorded in the month of April. The total contribution of the snow meltwaters and precipitation to the flow regime of the catchment area was 534 mm. The melt proceeded in four distinct steps: i) first melt, 189 mm, JD 85-106; ii) intervening cold period, JD 106-116; iii) gradual rewarming, 19 mm, JD 116-123; iv) main melt, 323 mm, JD 124-139. The hydrograph (Figure 2) shows small rising limbs on April 5 (JD 95) and April 17 (JD 107). On May 3 (JD 124) a large surge in the rising limb of the hydrograph was accompanied by the appearance of numerous throughflow inputs mostly at the northeastern edge of the lakeshore which persisted for 11-12 days. The hydrograph oscillated between 7×10^3 and 10×10^3 m^3 per day until its gradual descent (JD 140-155) to near pre-melt values.

The ice cover lost 300-350 mm of meltwater during the first melt, remained stable during the cold period and finally melted completely (additional 400-450 mm of meltwater) between May 3 and ice-off on May 13 (JD 134). Melting of ice cover, however, is not directly reflected in lake discharge as the meltwaters entering the lake body only compensate for the loss in weight of the melted cover if the cover is floating. The relatively small amount of discharge actually generated by the melting ice is related to the degree of anchoring of ice on the shore line and the lift of the cover.

Lake water chemistry at the lake discharge

With the onset of the first melt period (JD 85-106) the chemical composition of lakewaters discharged at the lake outlet started to change (Figure 3). The melting of the snow on the ice cover and the diffusion of the snow and ice meltwaters into the upper sub-ice pre-melt waters resulted in a gradual decline of total cation concentrations accompanied by a decrease in conductivity and pH. On the other hand, concentration changes of the strong-acid anions were far smaller; there was a slight rise in total anion concentrations followed by a decline to just below pre-melt values during the same period.
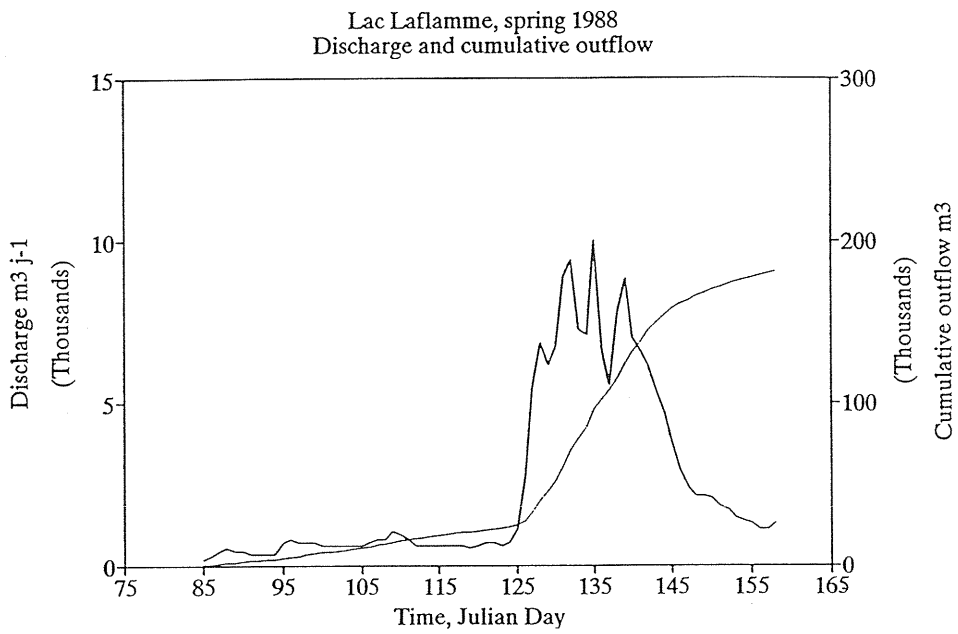
Lac Laflamme, spring 1988
Discharge and cumulative outflow

Figure 2. Hydrograph of the springmelt period at the lake discharge, Lac Laflamme, Québec, 1988.

———— Discharge   ———— Cumul. outflow
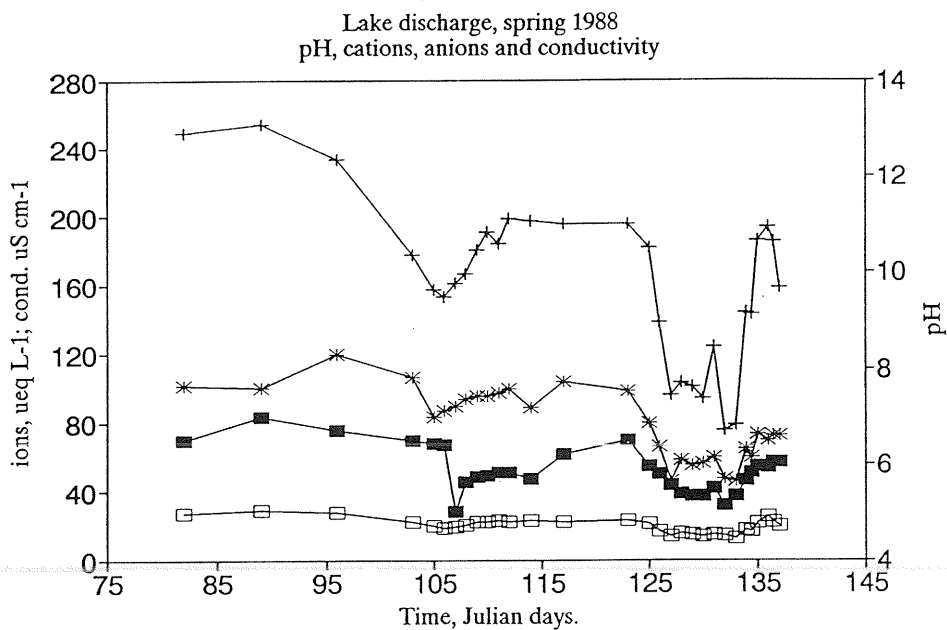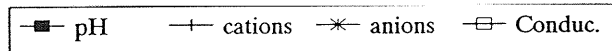
Lake discharge, spring 1988
pH, cations, anions and conductivity

Figure 3. Chemical composition of the lake discharge waters during the springmelt period, Lac Laflamme, Québec, 1988.

—■— pH   —+— cations   —*— anions   —□— Conduc.

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During the following cold period when meltwater inputs to the lake ceased, cation and anion concentrations, conductivity and pH generally increased towards pre-melt values; there was, however, a striking anomaly at the beginning of the cold period when a sharp significant drop in pH (6.4 to 5.0; JD 107) accompanied the initial increases in cation and anion concentrations and conductivity. During the main melt (JD 124-139), both cation and anion concentrations dropped appreciably; ion concentrations, pH and conductivity followed the same pattern.

Pre-melt lakewater and groundwaters are very similar in chemical composition; we can thus represent the overall chemical evolution of the lakewaters during the two melt periods by a simple two-component model. The first component is snow and ice-cover meltwaters or meltwater-induced runoff (throughflow) and the second is the pre-melt lakewaters. Chemical analysis of the different waters shows that the first component is characterized by relatively low conductivity, low cation concentrations and variable anion concentrations. Snow contains very low quantities of cations but relatively high concentrations of the strong-acid anions (SO_4 and NO_3). Initial snow meltwaters show high anion concentrations due to leaching of snow during meltwater percolation while later meltwater discharges contain low concentrations (Tranter, 1991). The second component consists of higher conductivity waters with high concentrations of cations and variable anion concentrations. These pre-melt lakewaters show SO_4 concentrations very similar to those of initial snow meltwaters while concentrations of NO_3, although generally low in lakewater, can vary significantly. Mixing of the two components within the lake results in a dilution of base cation concentrations of the pre-lake waters while the effect on anion concentrations may result in the dilution of either component depending on the chronology of meltwater production.

Conductivity values can generally be regarded as a measure of dilution of ion concentrations. In the case of different lake-water samples at Lac Laflamme during the whole melt period the relationship between the concentrations of base cations (Ca, Mg, Na, K) and conductivity (Figure 4; r^2 = 0.97) reflects the distinct differences in cation composition between the two components. In the case of the anions, however, the fact that mixing may cause dilution to either component results in a relationship between anion concentration and conductivity which is less significant (Figure 5; r^2 = 0.65). Closer analysis of the data shows that the relationship for SO_4 is far closer to the two component model (r^2 = 0.80) than that for NO_3 (r^2 = 0.04). The SO_4 concentrations in the lake during melt are more likely to reflect the same overall phenomenon of dilution as the cations; on the other hand, the behaviour of NO_3 seems to depend on other processes as well as dilution.

The dilution of cations has been shown to be a major factor in the pH depressions shown by surface waters of the Canadian Pre-cambrian Shield during the spring melt (Molot et al, 1989). In many cases anion dilution also occurs and contributes to the decrease in alkalinity (increase in acidity) of the waters. The gradual decrease in pH (6.8-6.4) during the first melt period (JD 85-106) may be attributed to base cation dilution caused by the mixing of snow and ice-cover meltwaters with the underlying pre- melt lakewaters; the initial increases in anion concentrations would tend to increase in acidity. Using the simple two-component model for lakewater chemistry we can estimate from the concentrations of total cations in the pre-melt lakewaters (260 μeq L^{-1}), in snow/ice-cover meltwaters (25-30 μeq L^{-1}) and the lake outlet (160 μeq L^{-1}) that the snow and ice-cover meltwater that entered the lake body mixed in with approximately 0.77 times its own volume of pre-melt lakewater (mixing ratio for lake:meltwater = 1:1.3) during its residence time in the lake. From the concentration of SO_4 in the pre-melt waters (90-100 μeq L^{-1}) and snow/ice-cover meltwaters (measured mean 30 μeq L^{-1}) the mixing ratio is 1:1. If we use the same method to calculate the mixing ratio for NO_3 (meltwaters, 15 μeq L^{-1}; pre-melt waters, 3-5 μeq L^{-1}) then the mixing ratio becomes 1:0.5). This would suggest that during the mixing of meltwater and pre-melt
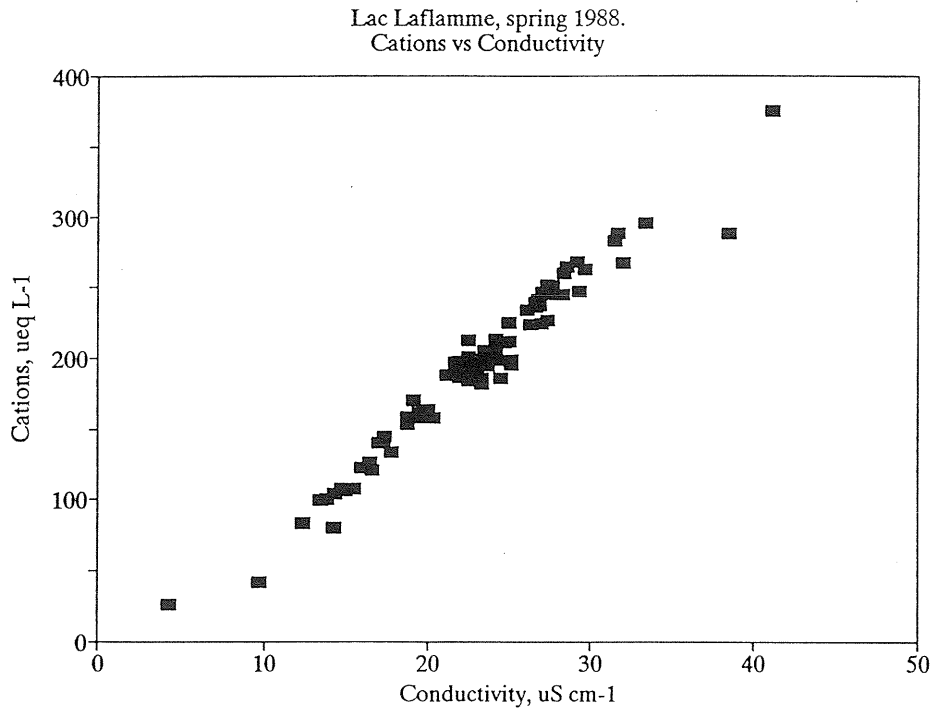
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Lac Laflamme, spring 1988.
Cations vs Conductivity

Figure 4. Base-cation concentrations and conductivity of lakewaters during the springmelt period, Lac Laflamme, Québec, 1988.

Lac laflamme, spring 1988.
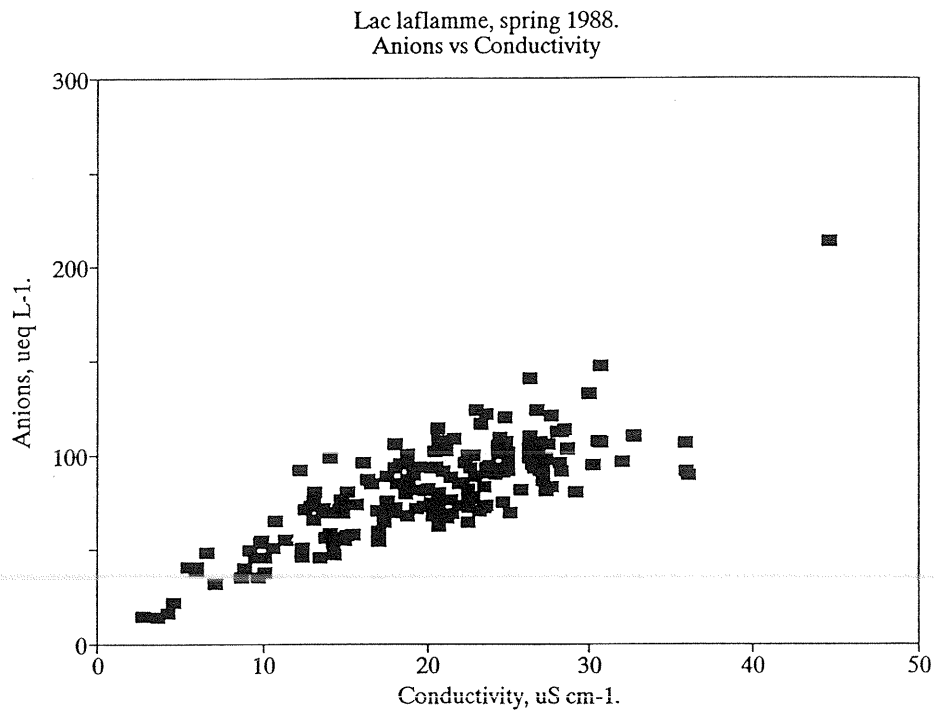Anions vs Conductivity

Figure 5. Anion concentrations and conductivity of lakewaters during the springmelt period, Lac Laflamme, Québec, 1988.

waters some NO_3 may have been removed from the system -presumably by biological activity. The mixing ratios are, however, very approximate as the chemical composition of snow and ice-cover meltwaters from the ice-cover varied greatly and not enough samples were taken during the study period to determine if the results are significant or not.

On JD 107 pH of the outlet waters dropped sharply from 6.4 to 5.0. Just prior to the pH depression both the dilution of cations and anions had resulted in low values for the concentrations of these ions. However, slight progressive increases in the ion concentrations of the discharge waters were evident at the time of the depression. The increase in the anion concentrations, however is more the result of a rise in the NO_3 concentrations rather than SO_4 (Figure 6). The divergent patterns of SO_4/NO_3 concentrations during the episode can be explained by the results of the rhodamine tracer experiments. Figure 7 shows the rhodamine concentrations at the lake discharge. The first pH depression occurs at the time that the rhodamine front from the injection of the tracer in the lakeshore snowcover (Figure 7, arrow A) reaches the lake outlet. From the date of the first tracer injection and the detection of rhodamine at the outlet, the experiment showed that during the initial melt period snow meltwaters from the shore had flowed onto the inshore ice, percolated down to the upper sub-ice layers and moved towards the lake outlet at an average velocity of 0.3 mm s^{-1} under laminar flow conditions (Roberge and Jones, 1991). The pH depression is thus the result of the arrival of this meltwater front containing relatively high concentrations of NO_3, at the lake outlet.

Typical concentrations of NO_3 in initial meltwaters from snow at Lac Laflamme are between 40 to 90 μeq L^{-1}, while those of SO_4 from 80 to 130 μeq L^{-1} (Jones and Bedard, 1987). The meltwater front moved through the upper layers of partially mixed ice-cover meltwaters and pre-melt lakewaters which were becoming progressively more diluted in cations and SO_4 as the front advanced. It is thus difficult to calculate a mixing ratio for the front. If we use, however, a mean approximate NO_3 concentration of 8 μeq L^{-1} for the ice-cover/pre-melt lakewater mix, the discharge of waters containing 25 μeq L^{-1} of NO_3 in the outlet waters during the pH depression suggest that the mixing ratio for the lakewater:meltwater front is between 1:2 and 1:3.

The second pH depression occurred during the main melt sequence (JD 124-139), immediately after the onset of throughflow inputs to the lake, when the pH declined again from a value of 6.4 (to which it had risen during the intervening cold period) to 5.2. In this latter episode, however, the decline was more gradual than in the case of the first pH depression. The second pH episode is again characterized by overall base cation and anion dilution except in the case of NO_3 which rose at the beginning of the event (JD 124-125) and then dropped to very low values during the main sequence. The initial rise in NO_3 concentrations is due to the melting of snow which had accumulated on the ice cover during the preceding cold period. The subsequent decrease in NO_3 is due to the arrival at the lake outlet of large influxes of surface runoff to the lake. The second rhodamine injection experiment (Figure 7, arrow B) showed that hillslope throughflow moved rapidly through the lake as a turbulent layer 20-40 cm thick at a velocity of 6 mm s^{-1}.

The second pH depression is thus initiated by new snow meltwaters from the ice-cover melt but is subsequently dominated by the acidic pulses generated largely from the quickflow of meltwaters through the upper (organic) soil horizons. Throughflow inputs to the lake are characterized by very low concentrations of NO_3 (0- 10 μeq L^{-1}); concentrations of SO_4 are approximately 60 μeq L^{-1}. During the main melt sequence the residual meltwaters from the ice cover become dilute and mixing of these meltwaters, lakewaters and the turbulent throughflow results in negligible concentrations of NO_3, decreases in SO_4 concentrations to 40 μeq L^{-1} and an increase of the pH of the throughflow from a mean value of 4.3 upon input to the lake to 5.2 at the lake outlet. The low values of NO_3 in throughflow is the result of two processes. In
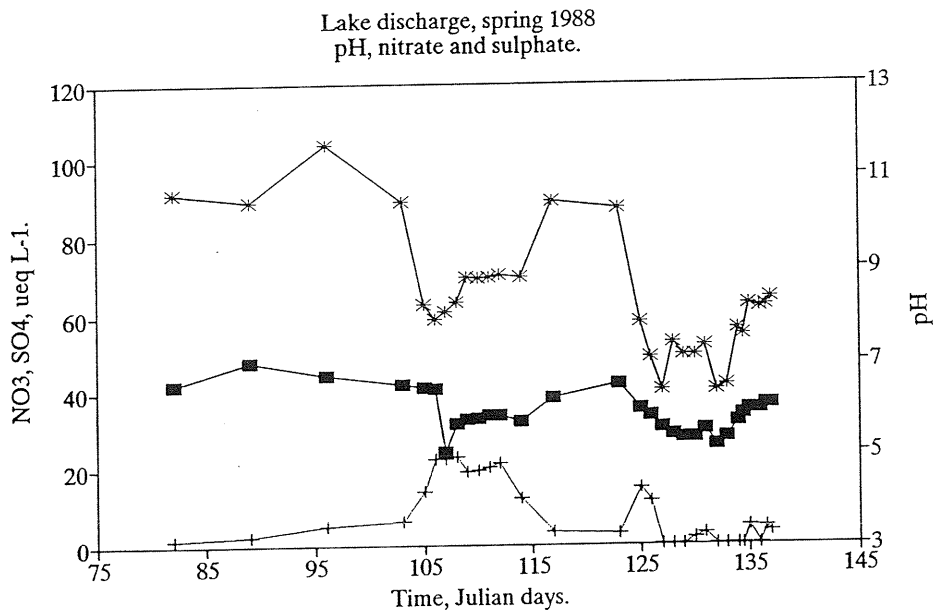
Lake discharge, spring 1988
pH, nitrate and sulphate.

Figure 6. pH, NO_3, and SO_4 concentrations of the lake discharge waters during the springmelt period, Lac Laflamme, Québec, 1988.

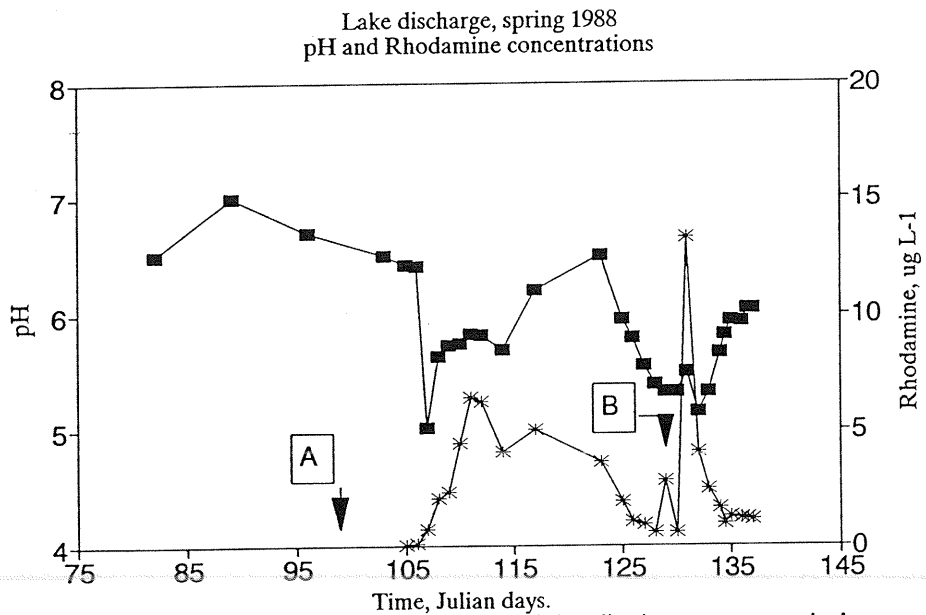Lake discharge, spring 1988
pH and Rhodamine concentrations

Figure 7. Rhodamine concentrations of the lake discharge waters during the springmelt period, Lac Laflamme, Québec, 1988: A, injection of shore snowbanks; B, injection of surface runoff

the first, the meltwaters which give rise to the runoff are dilute having lost most of their chemical load during the relatively small discharges of the early melt period. Secondly, NO_3 is transformed in the soil by biological activity; both NO_3 assimilation or denitrification could be involved (Hendershot et al, 1992).

During both pH depressions it can be seen that the behaviour of the strong-acid anions NO_3 and SO_4 may concur (dilution, main melt sequence) or diverge (dilution [SO_4] versus increases in concentration [NO_3], first pH episode). The results for SO_4 are similar to those of Molot et al (1989) who noted that during alkalinity depressions of shield surface waters in Ontario SO_4 dilution generally occurred, although some of the episodes did show an increase in SO_4 concentrations. The authors, however, noted that the overall effect of changes in NO_3 on alkalinity depressions were not judged to be significant, but do nevertheless comment on the observation that NO_3 can contribute to depressions in alkalinity of stream waters in early melt periods. Analysis of their data shows that some NO_3 increases are concomitant with SO_4 dilution as in the case of the first pH depression at Lac Laflamme.

Chemical profiles in the sub-ice waters
The chemical composition of lake discharge can only be regarded as a integration of the chemistry of all the different flowpath patterns that take place under the ice cover (Molot et al, 1990). The chemical profiles of the sub-ice waters, particularly near the shoreline, can reflect finer details regarding both the origins of inputs to the lake and sub-ice processes on a more local scale.

Figure 8 shows the evolution of pH, SO_4 and NO_3 in the sub-ice waters of the lake at the access hole B20. From the pH profile it can be seen that two distinct pH fronts passed through the area of B20 during JD 103-105 and from JD 125 on respectively; these were subsequently recorded at the lake outlet on JD 107 and from JD 126 onwards. The B20 SO_4 profile shows that the fronts are associated with low concentrations of this ion (45 μeq L^{-1}, first pH depression; 25 μeq $^{-1}$,

second depression). These values are lower than those for the lake outlet; the higher values at the lake discharge probably reflect increased mixing as the fronts move through the lake and the effect of groundwaters updrawn from the deeper layers at the outlet. The chemical gradients show the extent to which the meltwater fronts influence the chemistry of the lakewaters i.e. within the first 40 cm below the ice; this is in agreement with the rhodamine data for B20 (Roberge and Jones, 1991).

The NO_3 profile shows that the first pH front was characterized by high concentrations of NO_3 (30 μeq $^{-1}$) and the second by very low concentrations (5 μeq L^{-1}) of the ion. Just prior to the second depression there is an increase of NO_3 associated with the meltwater input from the melting snow on the lake. What distinguishes particularly the NO_3 profile from that of SO_4, however, is the pattern of NO_3 that evolves during the cold period between JD 113-119. The profile shows that there was a substantial decrease in NO_3 in the layer 10-20 cm below the ice surface. This was accompanied by an increase in the pH from 6.0 to 6.2. As concentrations above and below this particular layer showed relatively constant concentrations of the ion and as the sub-ice waters were fairly stagnant due to very low flows at the lake outlet, we attribute the decrease of NO_3 in the B20 area to biological activity by algae.

CONCLUSION

This study has demonstrated the origins of the meltwaters associated with episodic pH depressions in Lac laflamme during the springmelt. The work supports the study by Gunn and Keller (1986) who reported that melting snow from the lake surface can be a source of short early pH depressions. The results from Lac Laflamme, however, also indicate the greater importance of snow meltwaters along the lakeshore and of throughflow waters as acid inputs to the lake at different times of the melt period.

This study has further calculated the extent of mixing of meltwaters and pre-melt lakewaters
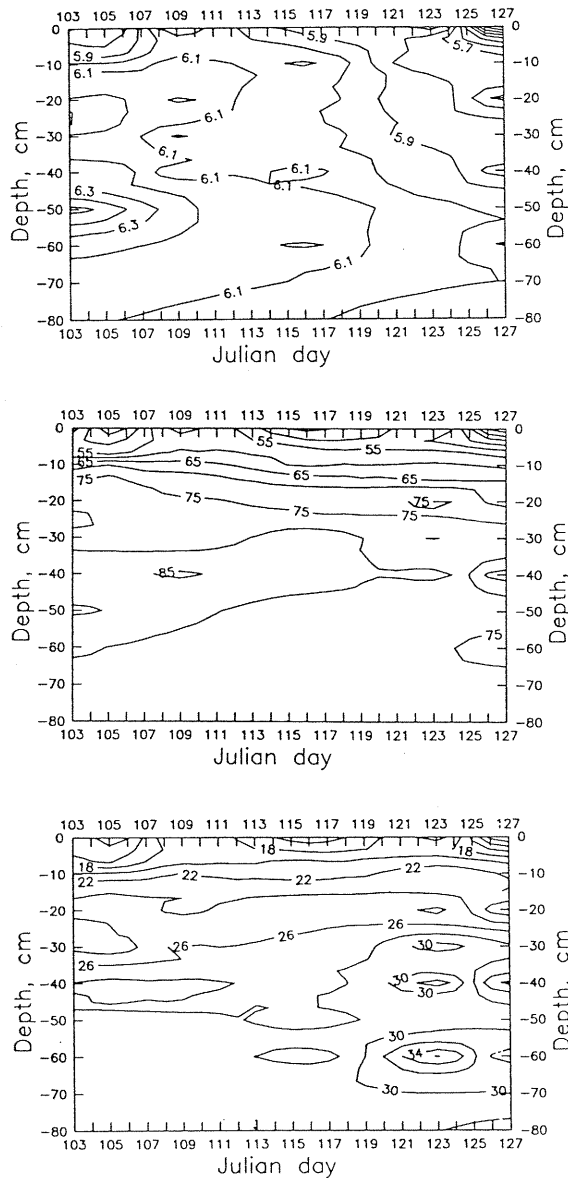
Figure 8. pH (top), SO₄ (middle), and NO₃ (bottom) concentration profiles at access-hole B20 during the springmelt period, Lac Laflamme, Québec, 1988.

from the chemical composition of the lake discharge and sub-ice waters. The results are similar to those of Bergmann and Welch (1985) who demonstrated by the use of rhodamine that meltwater mixing under ice cover is restricted to upper layers often only a few centimeters thick.

The Lac laflamme study has also shown that both cation and overall anion dilution (Molot et al, 1989) are responsible for the pH depressions. The concentrations of the strong-acid anions, SO_4 and NO_3, however, show either different or similar patterns depending on the origin of the acidic pulses.

The first pulse of acidity shows high concentrations of NO_3 and low concentrations of SO_4. The second pulse contains very low concentrations of both ions. The difference in these ionic patterns of the pulses reflect the chemical signatures of snow meltwaters and surface runoff. The removal of NO_3 from meltwaters routed to the lake through soil horizons may be due to denitrification in the soil and assimilation in the root zone during meltwater infiltration.

The study also suggests that NO_3 removal may also occur in sub-ice waters due to assimilation by phytoplankton.

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The presence of the ice cover accentuates the problem of episodic acidification of running waters downstream of the lake. The passage of acid inputs through the lake are confined to shallow layers under the ice, mixing is restricted and neutralization by pre-melt lakewaters with high alkalinity is low.

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