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#### ABSTRACT

Mass balances of  $NO_3$  and  $NH_4$  in meltwater from the snowpack, in runoff and in subsurface waters in a Boreal forest catchment (Lac Laflamme, Québec) were calculated for the 1985 spring melt period. The results demonstrated that depletion of both ions occurs in the snowpack when the free water content of the pack is high. This is believed to be due to microbiological activity. However, over the whole melt period (27.03 - 16.05), the preponderance of dry deposition to the pack over in-pack microbiological activity during days of low meltwater discharge resulted in a net increase of  $NO_3$  (0.83 meq m<sup>-2</sup>) over and above the sum (11.74 meq m<sup>-2</sup>) of the amount stocked in the original snowcover before melt (7.46 meq m<sup>-2</sup>) and the total load deposited by wet precipitation (4.01 meq m<sup>-2</sup>) during melt. In the case of  $NH_4$ , a similar mass balance calculation showed that the net result of the above processes was a  $NH_4$  depletion (1.03 meq m<sup>-2</sup>) in the pack.

For the combined melt period and forest-floor runoff period (27.03 - 30.05), the mass balance calculations for  $NO_3$  and  $NH_4$  in runoff per unit area of a gauged small catchment indicated that the soil-vegatative system acts as a sink for both the ions (1.3 meq m<sup>-2</sup>,  $NO_3$ ; 4.66 meq m<sup>-2</sup>  $NH_4$ ) during the period. The immobilisation of nitrogen in the soil during the forest floor runoff regime represents a critical phase in the nutrient cycle of boreal-forest ecosystems. At this time, the rate of hydrological transfer of these nutrients to the forest floor attains its maximal value.

## INTRODUCTION

Recently, Johnson et al. (1986) concluded that although acidic pollutants deposited from the atmosphere  $(SO_2, SO_4, NO_x)$  are not the only cause of spruce die-back in the Adirondacks, they may play a significant role by weakening the capacity of trees to survive severe natural conditions. The importance of this role can only increase if emissions of such pollutants are maintained or continue to rise over the next few decades. Although no such damage has been reported in the case of the boreal forest in eastern Canada (Morrison, 1984), the die-back in the Adirondacks could well be a presager of future conditions for a resource which is the principal component of the economy of Québec and the Maritime Provinces. In a similar vein, Nihlgard (1985) has hypothesised that the combined effect of acid deposition and over-supply of nitrogen by  $NO_3$  and  $NH_4$  is responsible for the long-term tree damage observed in Europe. In order to establish background data for the determination of the hydrological and biogeochemical control of nitrogen fluxes in boreal forest systems, we have, since 1983, studied the evolution of  $NO_3$  and  $Nd_4$  in the Lac Laflamme watershed north of Québec City. The studies have mostly been concerned with the spring-melt period. At this time, the forest floor experiences a severe hydrological and chemical discharge from the snowpack (Johannessen and Henrickson, 1978). The snowpack can accumulate up to 20-40% of the annual wet deposition load of pollutants before melt (Jones and Deblois, 1987), while streamflow over the whole melt period can represent 34% of the annual flow (Papineau, 1985).

The present article reports the results of a mass-balance study on the fluxes of  $NO_3$  and  $NH_4^+$  through the snowcover, in the forest floor runoff and in the subsurface waters of the Lac Laflamme watershed during the spring melt period of 1985.

#### SITE DESCRIPTION

The watershed of Lac Laflamme (0.684 km²; lake basin, 0.061 km²) is situated (46°11'N, 74°57'W) 80 km north of Québec City. Mean annual temperature is 0.2°C (mean minimum -15°C, January; mean maximum 15°C, July). Annual precipitation is 1 400 mm of which 490 mm fall as snow (Plamondon, 1982). The water equivalent of the snowcover before the main melt period varies from 300 mm to 400 mm (Jones, 1987). Balsam fir is the predominant species, black spruce and white birch are present in small amounts. A humic podzol covers a thick layer of till and moraine, which in turn overlays the precambrian gneiss of the Grenville Formation.

The hydrographic network is very poorly developed with only one main tributary; this enters the lake via a marsh. During the spring-melt period, intermittent streams (which run in depressions in the organic soil) constitute the forest floor runoff component to the permanent stream channel or lakeshore seepage zones. These streams are fed by lateral flow (including pipeflow) through the organic horizons and also by resurgence of subsurface waters. The latter arise from the lower soil horizons up through the organic horizons when the water table is high (Roberge & Plamondon, 1987). The intermittent streams can change flow direction with flowrate. They can only be reasonably gauged in well defined gullies where flow direction becomes independent of flowrate.

#### METHODOLOGY

Precipitation (snow and rain) was collected by means of two wet-only collectors (Sangamo Type A). One collector was placed in an open clearing, while the other was installed under a tree canopy (balsam fir). The volume of precipitation was measured either directly in the collector (rain) or after melting snow in the laboratory.

The water equivalent (WE) of the snowcover was determined by weighing snowcores taken with the use of a Adirondack-type corer (Gamma, Hempstead, NY). Snow meltwater was collected by three lysimeters (fibreglass tanks,  $1 \text{ m} \times 1 \text{ m} \times 0.5 \text{ m}$  height). One lysimeter was placed in an open clearing, one under a balsam fir-white birch open canopy and the third was installed under a close-knit canopy of balsam fir only (details of lysimeter construction may be found in Jones and Deblois, 1987).

Forest floor runoff was collected as grab samples in plastic bottles at a temporary gauging station that was installed to measure the flowrate of an intermittent stream. Subsurface waters (0-1 m depth) were pumped up to ground level through a piezometer tube by means of a peristaltic pump. Due to the design of the piezometer tube (Jones and Bédard, 1987), samples are more representative of the chemical composition of an integrated sample over the depth of the saturated zone than of the water quality at any one level.

# Sampling frequency

The study period started on March 28 (1st melt) and finished on May 30 (cessation of forest-floor runoff). Sampling of precipitation was carried out on an event basis. If an event extended over 24 hrs the collectors were sampled every 24 hrs. Snow cores were taken every 24 hrs. Lysimeters, runoff, and subsurface waters were collected every 24 hrs when discharge and/or flow over the forest floor or the presence of water in the piezometer tube warranted collection.

# Chemical analysis

All liquid samples for  $NO_3^-$  were analysed within 48 hours of sampling (conservation

temperature 4°C). Samples for NH $_{4}^{+}$  analysis were conserved with H $_{2}$ SO $_{4}$  (1-2 weeks) until analysed. Solid samples were kept at -20°C until melted. The samples were filtered (0.4  $\mu$ m nucleopore) and the analyses were carried out using a Dionex 12S ion chromatography unit (NO $_{3}$ ) and colorimetry (indophenol-sodium nitroprusside; NH $_{4}^{+}$ ).

## Data analysis

All calculations of NO $_3$  and NH $_4^+$  loads either deposited by precipitation (including canopy throughfall, P<sub>L</sub>), stocked in the snowcover (S<sub>L</sub>), discharged by lysimeters (L<sub>L</sub>) or present in runoff (R<sub>L</sub>) were converted to the common unit of meq m<sup>-2</sup> (equation 1)

$$\begin{array}{lll} P_{L} &=& \begin{bmatrix} C_{P} \\ S_{L} \\ \end{bmatrix} & V_{P} & / & A_{P} \\ V_{S} & / & A_{S} \\ L_{L} &=& \begin{bmatrix} C_{L} \\ C_{L} \\ \end{bmatrix} & V_{L} & / & A_{L} \\ V_{R} & / & A_{R} \end{array} \tag{1a} \tag{1b}$$

- •• where  $[C_p]$ ,  $[C_S]$ ,  $[C_L]$  and  $[C_R]$  are the concentrations of the ion (NO $_3$ , NH $_4$ ,  $\mu$ eq L $^{-1}$  = meq m $^{-3}$ ) in precipitation, snowcover, lysimeter discharge and runoff respectively.
- ••• where  $V_P$ ,  $V_S$ ,  $V_L$  and  $V_R$  represent the volume (m $^3$ ) of precipitation collected by the wet-only collector, the volume of snow in the snowcover, the volume of lysimeter discharge and the volume of runoff respectively.
- •• where  $A_p$ ,  $A_S$ , are the interior cross-sectional area (m²) of the wet-only precipitation collector and the interior cross-sectional area of the snow corer  $A_L$  and  $A_R$  are the catchment area of the lysimeter and the catchment area of the forest-floor runoff respectively.

The catchment area of lysimeters varies with evolving flow patterns in the snowcover (Marsh and Woo, 1985). The true catchment areas of the lysimeters for any one period (> 24 hrs) were determined hydrologically by comparing the amount of water discharged by the lysimeters to the sum of snowcover depletion and added precipitation (Jones and Deblois, 1987).

The determination of the catchment area of the runoff plot is more complex. This is due, in part, to the fact that the origin of the flow evolves with the progression of the variable source area (VSA, Roberge and Plamondon, 1987). VSA itself depends to a large extent on the level of the water table. In addition, the time lag between the maximal flow of forest runoff and that of the meltwater lysimeter varies from 4-10 hours. It therefore becomes difficult to directly compare the hydrogeochemical fluxes from the lysimeters with those recorded in the forest floor runoff.

We thus calculated a mean value for the catchment area of the intermittent stream for the period that the forest floor runoff actually occurred (25.04 - 30.05). The calculations were achieved by developing 4 models in which  $A_R$  is a parameter (Jones and Bédard, 1987). Three of the models were hydrological; (models 1, 2, 3; Table 1); they relate the area of the watershed, the volume of the lake or the level of the water table, and the total flow at the lake outlet to the total flow of the intermittent stream for the whole runoff period. The catchment of the latter was then calculated by direct proportionality. The fourth model is a hydrochemical model. It assumes that the Cl<sup>-</sup> ion is conservative in the forest system during the snow melt period. The catchment area of the forest runoff may then be calculated from a balanced Cl<sup>-</sup> budget, where the total Cl<sup>-</sup> load exported by runoff is derived exclusively from the total load discharged by the lysimeters (Jones and Bédard, 1987). Table 1 shows the essential methodology for the data analysis and the values for the catchment areas of each component of the precipitation-lysimeter-runoff systems.

In the case of groundwater, we do not have enough data collection sites to attempt to calculate the increases or decreases of  $NO_3^-$  or  $NH_4^+$  load (meq m<sup>-2</sup>) in the subsurface waters as the water table rises or redescends above the pre-melt level.

Table 1 Method of calculation of NO $_3$ , NH $_4^+$  loads meq m $^{-2}$  in components of a boreal forest ecosystem (Lac Laflamme, Québec) during the spring melt period, 1985

Sampling method (number of units in operation)	Catchment area, m <sup>2</sup>	Calculated load, meg m <sup>-2</sup> (24 hr intervals) for melt period (28.03 - 16.05)	Calculated load for whole melt-runoff period (28.03 - 30.05)
Wet-only collectors (2)	0.31	$[C_P]$ $V_P$ / 0.31	$\Sigma_{28.03}^{30.05}$ ([ $c_{ m p}$ ] $v_{ m p}$ ), / 0.31
Corer (Adirondack)	80.0	[c <sub>S</sub> ] v <sub>S</sub> / 0.08	$\Sigma_{28.03}^{16.05} ([c_{\rm S}] v_{\rm S})_{i} / 1.07$
Lysimeters (3)	0.79 - 1.39 mean 1.07 ± 0.30	$\left[ c_{\mathrm{L}} \right]  \mathrm{V}_{\mathrm{L}}  /  1.07$	$\Sigma_{28.03}^{16.05}$ ([ $c_{ m L}$ ] $v_{ m L}$ ) $_{ m i}$ / 1.07
	Model 1, 1741 Model 2, 1712 Model 3, 1513 Model GI, 1244 mean 1489 ± 234		$\Sigma_{25.04}^{30.05}$ [c <sub>R</sub> ] <sub>i</sub> + [c <sub>R</sub> ] <sub>i+1</sub> * $\Sigma_{i}^{i+1}$ v <sub>L</sub> **/ 1489
Piezometer			

\*  $\begin{bmatrix} C_R \end{bmatrix}_{i+1}$  = concentration of ion in grab sample on day i  $\begin{bmatrix} C_R \end{bmatrix}_{i+1}$  = concentration of ion in grab samble on day i+1

\*\*  $\Sigma_{i}^{i+1}$  = total volume of flux in runoff from day i to day i+1

#### RESULTS AND DISCUSSION

Figure 1 illustrates the fluxes of nitrogen through the boreal forest system and the relationship between the measured loads for the specified time intervals in each component (precipitation, snowcover, lysimeter discharge, and runoff). Table II a) records the result of the mass balance calculation for the atmosphere-snow system. It shows the calculated fluxes of  $NO_3^-$  and  $NH_4^+$  in precipitation, snowcover and lysimeter discharge for the period from the  $1^{\rm St}$  melt (28.03) to the disappearance of the snowpack (16.05). Table II b) records the result of the mass balance calculation for the soil-runoff system. It shows the calculated fluxes for these ions in the precipitation for the period between the end of snowmelt and the cessation of streamflow, in the lysimeter discharge during the melt period and in the forest floor runoff during the period that flow was registered.

## Nitrogen mass balance for the atmosphere-snow system (precipitation, snowcover, meltwater)

The overall result of the mass balance for the atmosphere-snow system shows that the boreal forest snowpack is a milieu in which the net total nitrogen depletion takes place to the extent of 0.20 meq m<sup>-2</sup> (NH $^+_4$ , 1,03 meq m<sup>-2</sup> loss; NO $^-_3$ , 0.83 meq m<sup>-2</sup> gain). The total amount of nitrogen loss, however, cannot be calculated. We have no independant means for the calculation of the amount of nitrogen which is dry deposited on the snowcover. This would include gaseous (NH $^-_3$ , HNO $^-_3$ , NO $^-_2$ ), aerosols (NH $^+_4$ , NO $^-_3$ ) and particulates (NH $^+_4$ , NO $^-_3$  adsorbed, N-organic; Cadle et al., 1985).

Based on the results for the dry deposition velocities of nitrogen species published by Cadle et al. (1985) and the reported (or estimated) mean-concentrations of the same nitrogen compounds in the atmosphere at Lac Laflamme during the winter-spring season (Barrie and Sirois, 1986), we have estimated that the total amount of dry-deposited nitrogen from the atmosphere to be 0.27 meq m $^{-2}$  for the period 28.03-16.05 (Table III). This method, however, underestimates the total N that is dry deposited. It does not take into account all nitrogen species (eg. PAN) and, more importantly, does not consider litter fall (plus adsorbed aerosols) that occurs in relatively large quantities ( $^{\sim}$  167 m $^{-2}$ , Jones and Deblois, 1987) during the whole winter.

The calculated total N dry deposition (NO $_3$ , NH $_4$  and HNO $_3$ ) is minimal (0.27 meq m $^{-2}$ ) for the period 28.03-16.05. The resulting recalculation for total N depletion by microbiological activity is then 0.47 meq $^{-2}$  (0.20 + 0.27) and shows that biological activity and N-dry deposition are processes with comparable rates. However, both rates are low compared to those of the dominant processes of N gain and loss from the system i.e. wet precipitation (Lp, 7.66, meq m $^{-2}$ ) and meltwater discharge respectively (L $_1$ , 17.0 meq m $^{-2}$ ) during the same period. Melt experiments in the laboratory by Jones and Deblois (1987) showed that depletion of NO $_3$  and NH $_4$  did take place in melting snow to which litter from the forest canopy had been added. In a further series of experiments (unpublished) they found that this organic debris retained a quantity of the  $^{1.5}$ NO $_3$  which had been added to the snow prior to melt. They conclude that the immobilization of N is due to microbiological activity. Barry and Price (1987) have also reported nitrogen losses in meltwaters which were in contact with the forest flow by comparing nitrogen loads discharged from lined lysimeters and unlined lysimeters in a deciduous forest plot. They ascribed the nitrogen deficit to the presence of organic matter without, specifically mentioning microbiological activity as such.

Organisms responsible for microbiological activity in snow may include the micro-epiphytes, lichens, algae, bacteria and fungi associated with the organic debris deposited during the winter season (Jones and Sochanska, 1985). The ability of snowpacks to support populations of microorganisms is well known (Stein and Amundsen, 1967; Visser, 1973). As an example, Hoham and Mohn (1985) have described the growth characteristics of autotrophic cryophilic flagellates which can deplete nutrients in snowpacks during the zoospore stage of the life cycle.

## Nitrogen wass balance for the soil-runoff system (meltwater, runoff)

In an analogous manner to Table IIa), Table IIb) shows the mass balance calculation for the soil-runoff system during the melt-runoff period (28.03-30.05). Approximately 33.6% (6.02/17.92) of the sum of the total nitrogen discharged by the lysimeters during the melt

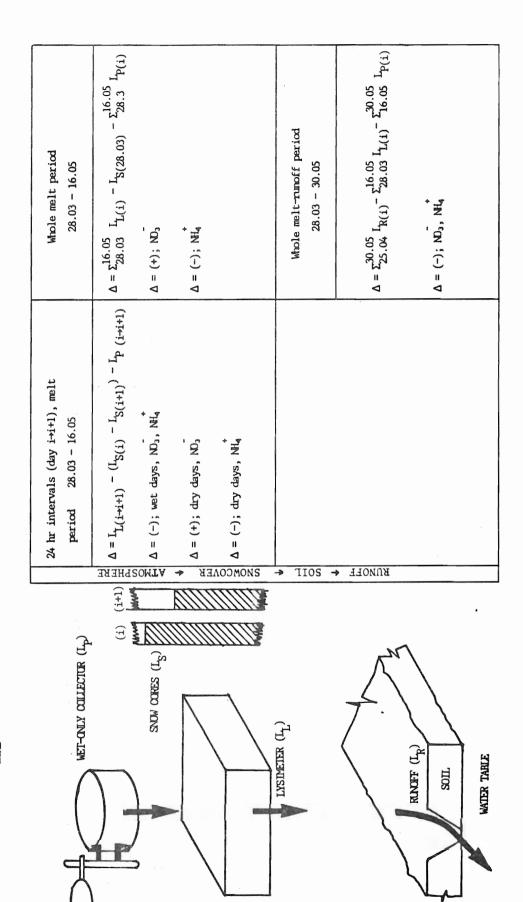


Figure 1: N Fluxes and mass balances calculations for snowmelt discharge and forest floor runoff in a boreal forest.

Table IIa)

Mass balance calculations for the atmosphere-snow system during the melt period (28.03 - 16.05), Lac Laflamme, Québec, 1985

Comment	Calculated flux (meq $m^{-2}$ ; 28.03 - 16.05)			
Component	NO <sub>3</sub>	NH <sub>4</sub>	Total N	
Precipitation (L <sub>P</sub> )	4.01	3.65	7.66	
Snowcover (28.03) (L <sub>S</sub> )	7.46	2.08	9.54	
Lysimeter meltwater ( ${ m L_L}$ )	12.30	4.70	17.00	
Mass Balance				
L <sub>L</sub> - L <sub>S</sub> - L <sub>P</sub>	0.83	(1.03)*	(0.20)	

# \* ( ), negative

Table IIb

Mass balance calculations for the soil-runoff system
during the melt-runoff period (28.03 - 30.05), Lac Laflamme, Québec, 1985

Component	Calculated flux (meq m <sup>-2</sup> ; 28.03 - 30.05)		
Component	NO <sub>3</sub>	NH <sub>4</sub>	Total N
Precipitation (16.05 - 30.05, L <sub>p</sub> )	0.60	0.32	0.92
Lysimeter (23.03 - 16.05, L <sub>L</sub> )	12.30	4.70	17.00
Runoff (25.04 - 30.05, L <sub>R</sub> )	11.60	0.30	11.90
Mass Balance			
$L_R - L_L - L_P$	(1.3)	(4.72)	(6.02)

Table III

AUTHORS	FORESTED CATCHMENT	NITROGEN SPECIES ( )	MASS BALANCE CALCULATION FOR CATCHMENT ( <u>MEA</u> N meq <sup>-2</sup> )	
Hemond & Eshleman, 1984	Mixed coniferous- deciduous	NO <sub>3</sub>	29.0 (R,yr <sup>-1</sup> )	
Hauhs, 1986	Coniferous	NO <sub>3</sub>	3.3 ± 9.5 (R,E)	
Likens <u>et al.,</u> 1987	Deciduous	NO <sub>3</sub>	42.1 ± 44.3 (R,E)	
Knight, 1985	Coniferous	NO <sub>3</sub> +N-Kjeldahl	1.4 ± 0.9 (R)	
Galloway, 1986	Deciduous	NO <sub>3</sub>	3.2 ± 4.2 (R,E)	
Bedard and Jones, 1987	Coniferous	NO <sub>3</sub> (1984) NO <sup>3</sup> + NH <sup>4</sup> (1985)	2.0 (E) 6.0 (R)	
R= Retention, E= Export, R,E= Retention or Export depending on year.				

period (17.00 meq  $m^{-2}$ ) and that subsequently deposited by precipitation after the end of the melt period (0.92 meq  $m^{-2}$ ) is retained by the soil systems while the water table is high and forest-floor runoff occurs.

Retention of nitrogen ( $NO_3$  or  $NH_4$ ) by forested basins on an annual basis in the North American Northeast is well known (Hemond and Eshleman 1984). From input – output budgets retention of  $NO_3$  may be relatively complete (20-100%, Hemond and Eshleman, 1984) or, on the other hand, there may be a net export of  $NO_3$  from forest systems by streamwaters. Likens et al (1977) have demonstrated the irregularity of  $NO_3$  mass-balances for a forest catchment area in New England, while Hauhs (1986) has also observed a similar behavior for a Norway Spruce stand in West Germany.

There are few nitrogen budgets from field studies of periods shorter than a year. Knight et al. (1985) concluded that N ( $NO_3$  + Kjeldahl N) is retained even during high hydrological flux in spring in a western lodgepole pine forest. However, Peters and Driscoll (1987) have observed exactly the opposite effect in a northern hardwood forest. These authors attributed NO3 release from the soil horizons into the meltwaters percolating through the forest flow to nitrification. In 1984 Bédard and Jones (1986) also found that there was a net export of NO2 from Lac Laflamme soil horizons into the forest floor runoff during the spring melt. NH4 was retained by the soil (Jones, unpublished). The data base, however, was very sparse and error bars on the calculation were much higher than those for similar calculations during the melt of 1985 (this work). Thomassin (unpublished results) has studied the interaction between meltwater and the upper soil horizon at Lac Laflamme and has concluded that denitrification is the dominant microbiological activity during the infiltration of meltwater into this horizon during the springmelt. Galloway et al. (1986) have calculated input (wet and dry deposition) - output (surface water) budgets for NO3 during the melt period for the catchment areas of lake systems in the hardwood forest region of the Adirondack mountains, NY. Depending on the lake basin studied and the year, basins may retain (1-3 meq m<sup>-2</sup>) or export (7 meq m<sup>-2</sup>) NO $_3$  during the spring melt.

To find any coherent pattern for nitrogen dynamics in these systems during the spring melt period is difficult with the limited data that is available on systems which themselves are extremely complex. It can be seen, for example that from the standard variations about the mean values for the annual N retention of the same system (Hauhs, Likens et al, Table III) fluxes can vary greatly from year. In some years net exports are recorded. We do know, however, that the result of nitrogen mass balances are the net result of the many processes occurring in the forest floor, and, in the case of forested basins with lakes and streams, processes within the lakes and stream themselves. From a consideration of the available data we can suggest a possible overall scheme for nitrogen dynamics during the snowmelt period. First,  $NO_3$  and  $NH_4$  may be leached from the forest floor in the initial stages of They are then reincorporated in the vegetative root zone as the net result of N immobilization by roots, micorrhizae, denitrifiers, etc. and adsorption by ion exchange as the melt progresses (Vitousek et al., 1982). This root zone is concentrated in the lower part of the organic layer (Vogt et al., 1983). If the rate of hydrological flux through this zone is faster than the net rate of N-assimilation.  $NO_3$  or  $NH_4^+$  will penetrate down to the mineral soil layers. Nitrogen species can then be routed to surface waters. Net export of N from the upper soil horizons during spring may well be determined by both the time and the magnitude of the hydrological flux during the melt.

## Nitrogen fluxes through the whole watershed

The results of the 1985 spring melt study show that of the total amount of inorganic nitrogen above ground level in the pack at the start-of melt and that added in precipitation during melt,  $0.20~{\rm meq}~{\rm m}^{-2}$  was immobilised in the snowpack (precipitation - snow - meltwater system) and  $6.02~{\rm meq}~{\rm m}^{-2}$  was further retained in the soil (meltwater - runoff system). It would thus appear that the role of the snowpack in the control of nitrogen fluxes is minor compared with that of the soil horizons. Even though, intuitively, we accept this as a true reflection of the N dynamics in the forest during the melt period, we would add a note of caution on comparing the two systems (snow  $\underline{\mathbf{v}}$  soil). In the case of the precipitation-snow-meltwater system, the hydrological data needed to calculate N fluxes in the system were relatively easy to obtain. The hydrological

characteristics of this system are well defined and hydrological inputs and outputs are directly related. Confidence in the data thus high.

Confidence in the measured values for hydrological data in the meltwater-runoff system is also high due to the use of the lysimeters and the gauged streams but, in this case, inputs cannot be directly related to output. The basic premise of the hydrological models in the calculation of the area of the forest floor which contributes to the gauged surface runoff is proportionality. It states that when the intermittent streams run their catchment area is of the same proportion to the area of the whole watershed as their flow is to that of the whole watershed (lake outlet discharge). This may be partially true on some days of high infiltration rates when the water table is high. It is certainly not true over the whole forest-floor runoff period (Roberge and Plamondon, 1987) when subsurface seepage to the lake also occurs. The surface area of the stream catchment area is overestimated and, as a consequence, mass balance calculations (Fig. 1) may underestimate the flux of N, mq m $^{-2}$  in the forest-floor runoff. Finally, all the hydrologic pathways in the catchment are not known. What appears in the stream bed on the rising limb of the hydrograph may have no direct relationship with the waters which infiltrated the soil just prior to the increase in stream flow. The presence of "old" or premelt water forced into streambeds by the infiltration of new meltwaters is well known (Obradovic and Sklash, 1987).

We are now carrying out studies in on both soil-lysimeters and surface waters at Lac Laflamme in order to elucidate the hydrological pathways in the watershed during the spring melt period.

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