

VARIATIONS IN THE CHEMICAL COMPOSITION OF THE SNOWPACK
AND ASSOCIATED MELTWATERS IN CENTRAL ONTARIO

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ABSTRACT

The major ion composition (H^+ , SO_4^{2-} , NO_3^- , NH_4^+ , Ca^{2+} and Cl^-) of the snowpack near Sudbury and Muskoka-Haliburton in central Ontario has been investigated by collecting snow cores, bulk deposition samples and associated meltwaters during the winters of 1977 to 1981. Concentrations of ions in the snow cores were less variable than concentrations in precipitation samples. The concentration of most ions (except Cl^-) was lower in the snowpack compared to corresponding cumulative bulk deposition. Consideration of H^+ and $SO_4^{2-} + NO_3^-$ concentrations at different levels in the snowpack profile suggests that the ions are lost by migration into the underlying soils and this can occur even during periods where no significant melting has been observed. During the spring melt period of 1979, all six streams entering Harp Lake in Muskoka-Haliburton experienced pH depressions. Compared to pre-melt concentrations, H^+ increased from 74 to 580%. Peak H^+ concentrations coincided with peak discharge in five of six cases (unlike observations made for Norwegian streams) and with declining NO_3^- concentrations in three of six cases (unlike observations made for Adirondack streams). Consideration of $SO_4^{2-}:NO_3^-$ ratios suggests that sulphuric acid is the dominant acidifying strong acid at Muskoka-Haliburton.

INTRODUCTION

Atmospheric deposition of substances is important for lakes and forests in the Shield environment typical of central Ontario. For example, Scheider *et al.* (1979a) have shown that direct atmospheric deposition of nutrients and H^+ is an important component of the total input to dilute lakes while Abrahamsen (1980) suggests that the nitrogen deposition accompanying acidic precipitation will likely enhance tree growth in Shield terrains.

Collection and analysis of snow cores has been used as a simple method for estimating atmospheric deposition of substances in northwest Ontario (Barica and Armstrong 1971), Manitoba (Franzin *et al.* 1979), Norway (Elgmork *et al.* 1973, Hagen and Langeland 1973, Wright and Dovland 1978), northern Minnesota (Eisenreich *et al.* 1980), and remote locations such as Greenland (Herron *et al.* 1977) and Antarctica (Delmas *et al.* 1980). When using this methodology for estimating winter (or annual) atmospheric

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deposition, it is implicitly assumed that the snowpack has not experienced significant chemical alteration. Elgmork *et al.* (1973) studied laminated snow profiles in Norway and reported that the snowpack remained well preserved as long as temperatures were below 0°C. Similar findings were reported by Henriksen (1972). They suggested that snow sampling in late winter would give valuable information on the chemical composition of precipitation, over the entire winter period and thus large regional snow core surveys were performed in Norway from 1974 to 1976 (Wright and Dovland 1978).

Subsequent comparison of snow core concentrations with corresponding values from precipitation collectors showed that the snowpack, in fact, generally had lower concentrations for many parameters (Wright and Dovland 1978, Skatveit and Gjessing 1979). This discrepancy may reflect the disproportionate loss of ionic species (relative to water) from the snowpack during periods of melting. Johannessen and Henriksen (1978) reported that 50-80% of the pollutant load in the snowpack samples was removed with the first 30% of the water in laboratory and lysimeter studies. The sudden loss of ions from the snowpack during melt has been used to explain springtime pH depressions observed in streams and the surface waters of lakes in Norway (Johannessen *et al.* 1980), Sweden (Hultberg 1976), Ontario (Yan and Stokes 1978; Jeffries *et al.* 1979) and northeastern USA (Hendrey *et al.* 1980). Such reductions in pH may detrimentally affect aquatic biota.

We have collected snow core samples in central Ontario in 1977, '78, '79, and '81 with the purpose of determining the variability in snowpack composition and its comparability with snow from precipitation samplers. In order to assess the impact of the release of acids from the snowpack on lakes in central Ontario, we have studied the meltwater chemistry.

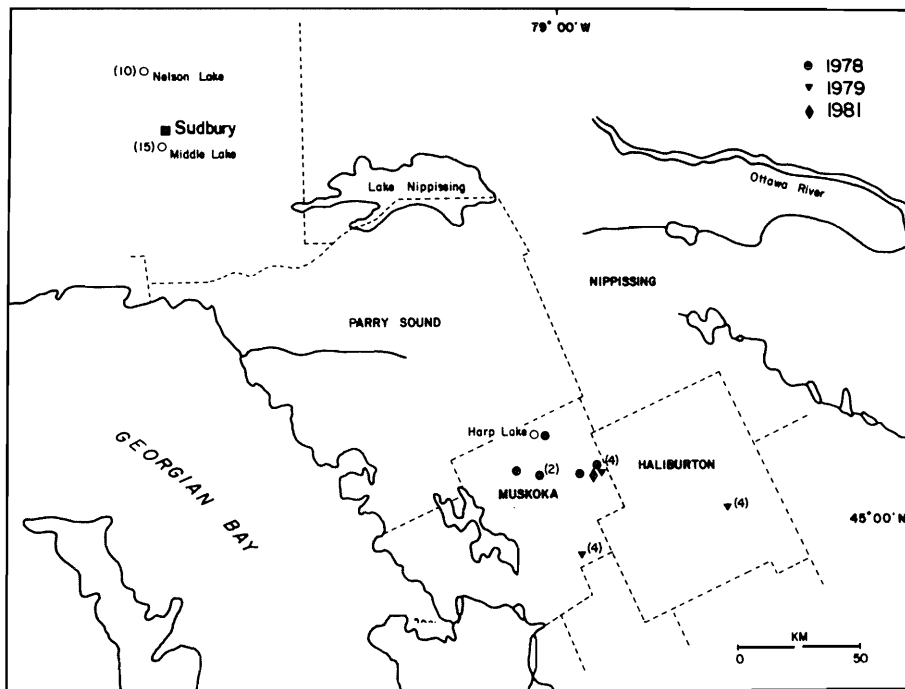


Figure 1. Location of snow core collections within the Muskoka-Haliburton and Sudbury regions of central Ontario. Snow cores at Sudbury (Middle and Nelson Lakes) were collected in 1977; year of collection at Muskoka-Haliburton is shown by an appropriate symbol. The number of cores collected at a location (when >1) is indicated by a superscript to the location symbol. The location of Harp Lake is also shown.

Study Area

The study area (Fig. 1) has been previously described (Jeffries and Snyder 1981, Scheider *et al.* 1979a). Snow cores were collected at two locations near the urban-industrial center at Sudbury and at one to six rural locations in Muskoka-Haliburton. The entire area is underlain by crystalline rocks of the Precambrian Canadian Shield and overlain by thin to absent glacially derived soils. The lakes of this area are considered potentially susceptible to acidification (Scheider *et al.* 1979b). The effect of snowmelt on Harp Lake was also studied. Harp Lake (Fig. 1) is a dilute (conductivity 30-40 $\mu\text{hos cm}^{-1}$), oligotrophic headwater lake with a surface area of 66.9 ha and total drainage basin area (including lake) of 530 ha. It is primarily used for recreational purposes.

Methods

Snow cores were obtained by pushing a clean cast acrylic tube down through the snowpack and then digging away the snow around the tube so that it could be retrieved with the contents intact. Care was taken to exclude any litter from the ground surface. Several cores obtained in this manner at a given site were melted and composited to provide sufficient volume for chemical analysis. All cores were taken in non-forested locations in order to eliminate potential "throughfall" effects. Johannes *et al.* (1980) have shown that snowpack storage of H^+ and NO_3^- is reduced under tree cover in Adirondack watersheds. On two occasions (Jan. 20, 1981 and Feb. 17, 1981) the snowpack at Muskoka-Haliburton was also sampled in profile by excavating a pit to ground level and taking a sample every 10 cm from the vertical face of the pit with a clean shovel. The weather prior to Jan 20, 1981 was extremely cold and no visible melting of the snowpack occurred. In the interval between Jan. 20 and Feb. 17, 1981, four days of warm weather occurred when maximum daily temperatures exceeded 0°C and, in fact, on Feb. 16 and 17 even minimum daily temperatures exceeded 0°C . During this period a rainfall of 38 mm was also recorded. The first profile sample therefore represents the "undisturbed" condition of the snowpack, while the second represents the condition after significant melting and influence by rainfall.

Stream chemistry was measured for the six major inflows to Harp Lake from February through May 1979. Samples were carefully collected at stream gauging structures in appropriately cleaned bottles to minimize contamination. Sampling frequency was roughly governed by discharge; thus prior to major melting, the streams were sampled weekly, while during periods of peak flow, samples were taken on a daily basis.

Snow cores were analyzed for pH, SO_4^{2-} , NO_3^- , Ca^{2+} , NH_4^+ , Cl^- and Al, stream samples for pH, SO_4^{2-} and NO_3^- . Chemical analyses of all water samples were completed as soon as possible after collection and always within 24 hr for non-conservative parameters. Analytical techniques are outlined in Ontario Ministry of Environment (1975).

Results and Discussion

The mean equivalent water depth and parameter concentration and percent coefficient of variation for snow cores collected near Sudbury (Middle and Nelson Lakes) and at Muskoka-Haliburton (Fig. 1) are given in Table 1. The data in Table 1 represent two different sampling scenarios. The Middle and Nelson samples were collected at prescribed intervals along a transect within an individual lake basin. Hence, although the samples represent a small area only, no attempt was made to eliminate variability introduced by local features (e.g., snow drifts, chemical influence of roads, etc.). On the other hand, the Muskoka-Haliburton samples were collected so as to be representative of the entire region, and therefore sampling of the snowpack at any obviously atypical location was avoided. Hence, variability in water equivalent depth was greater for the Sudbury area locations compared to Muskoka-Haliburton (Table 1).

Table 1. Mean water equivalent depth (cm) and concentration (H^+ , SO_4^{2-} , Ca^{2+} , Cl^- in $\mu eq L^{-1}$; Al in $\mu g L^{-1}$) for snow cores collected near Sudbury (Middle and Nelson) and Muskoka-Haliburton. The number of cores included in the mean and percent coefficient of variation is also given.

Parameter ¹	Middle			Nelson			Muskoka-Haliburton					
	Mean (n)	Coef. of Variation (%)	77 03 06	Mean (n)	Coef. of Variation (%)	77 03 06	Mean (n)	Coef. of Variation (%)	78 02 21	Mean (n)	Coef. of Variation (%)	79 02 21
Depth	54.4 (15)	19	35.6 (10)	33	15.7 (6)	8.6	25.5 (12)	13				
H^+	84 (15)	68	79 (10)	20	35 (6)	18	55 (12)	12				
SO_4^{2-}	56 (15)	23	42 (10)	22	19 (6)	25	19 (12)	12				
NO_3^-	45 (15)	33	52 (10)	8.7	35 (6)	13	37 (3)	5.6				
NH_4^+	24 (6)	16	17 (10)	18	19 (6)	13	7.9 (3)	9.1				
Ca^{2+}	26 (15)	20	29 (10)	34	12 (6)	35	16 (3)	41				
Cl^-	48 (15)	107	14 (10)	40	5.6 (6)	45	7.9 (6)	41				
Al	210 (15)	137	40 (10)	33	65 (6)	22	69 (11)	32				

¹ All parameters in $\mu eq L^{-1}$ except water equivalent depth (cm) and Al ($\mu g L^{-1}$)

The coefficient of variation for the snow core concentration ranged from 5.6 to 137% with only three cases (H^+ , Cl^- , Al at Middle) being $>45\%$. This degree of variation is small compared to the temporal variation in concentration observed either between individual precipitation events or between monthly cumulative precipitation samples. Ten-fold variation (or greater) in the composition of precipitation samples has been reported for both Muskoka-Haliburton (Scheider *et al.* 1979a) and Sudbury (Jeffries 1981). However, the snow core variability is generally consistent with the findings of Jeffries *et al.* (1978), Scheider *et al.* (1981) and Jeffries (1981) who reported no significant spatial variation in bulk deposition for all parameters in Table 1 (except Al) for regionally dispersed collectors at both Sudbury and Muskoka-Haliburton. Jeffries (1981) did note significant spatial variability in the bulk deposition of Al between collectors at and near Middle Lake and attributed this observation to the influence of nearby urban and industrial activities (no significant variability in Al was found at any other location). The larger coefficient of variation observed for H^+ , NO_3^- , and Cl^- concentrations at Middle compared to the other locations may also be attributed to local factors, particularly the presence of a road near the sampling site.

The mean concentration of H^+ , SO_4^{2-} , NO_3^- , Ca^{2+} and Cl^- are greater at the two Sudbury sites compared to Muskoka-Haliburton. Scheider *et al.* (1981) showed that there is no significant difference in the bulk deposition of either H^+ between Muskoka-Haliburton and any Sudbury location, or SO_4^{2-} between Muskoka-Haliburton and sites >12 km from the INCO smelting operations at Sudbury. Since the Middle Lake basin falls within this 12 km boundary, it is not surprising that elevated SO_4^{2-} concentrations are observed; however, since both H^+ and SO_4^{2-} are elevated at both Middle and Nelson (40 km from INCO), it is probable that there is significant dry deposition of SO_4^{2-} (or SO_2) to the snowpack which is not collected by a bulk deposition sampler. Jeffries *et al.* (1981) and Dillon (1981) have presented other independent evidence of significant SO_2 deposition at Sudbury. Greater snow concentrations of NO_3^- , Ca^{2+} and Cl^- probably reflect the greater vehicular density and utilization of road de-icing chemicals in the urban Sudbury area.

The relative concentration of H^+ , SO_4^{2-} , NO_3^- , Ca^{2+} , NH_4^+ and Cl^- in the snowpack compared to the corresponding bulk deposition is presented in Fig. 2. Of these parameters only Cl^- has approximately similar concentrations in both the snowpack and bulk deposition. With a few exceptions, the remaining parameters exhibited greater concentrations in bulk deposition in accord with the findings of Wright and Dovland (1978). Hydrogen ion concentrations in the snowpack ranged from 18% greater to 45% less than the corresponding bulk samples. Only the snow sample from the Middle Lake basin had a substantially greater H^+ concentration than the bulk deposition possibly due to "extra" dry deposition of SO_2 (not collected by the bulk sampler) at this near-smelter site. There was not a correspondingly greater SO_4^{2-} concentration in the snowpack however. The snow samples at Muskoka-Haliburton which exhibited the greatest absolute difference in H^+ concentration (compared to bulk) were all collected in the last half of February when short periods of melting were often observed. This evidence supports the observations of Johannessen and Henriksen (1978) for preferential loss of acids from the snowpack during the first melt.

Sulphate and NH_4^+ concentrations were always greater in bulk deposition (35 to 56% and 12 to 64% greater than the snowpack, respectively) while NO_3^- concentrations were either approximately equal (2 cases) or greater in bulk deposition (-13% to +36%). Calcium concentrations were always greater in bulk deposition (up to 61%) with one exception (Fig. 2).

It is clear from these observations that many chemical components of the snowpack are lost prior to spring melt and that this can occur even during extremely cold periods when there is no melting. Such factors as snowpack sublimation and dry deposition of pollutants should increase the snowpack concentration relative to bulk deposition rather than result in the above observations. Some chemical species show a greater degree of loss than others, notably SO_4^{2-} which is depleted more than either H^+ or NO_3^- .

Figure 3 shows the variation in H^+ and $SO_4^{2-} + NO_3^-$ concentrations in the snow profile in Muskoka-Haliburton in 1981. Prior to any melting (Jan. 20), the profile

SNOW CORES AND CORRESPONDING BULK DEPOSITION

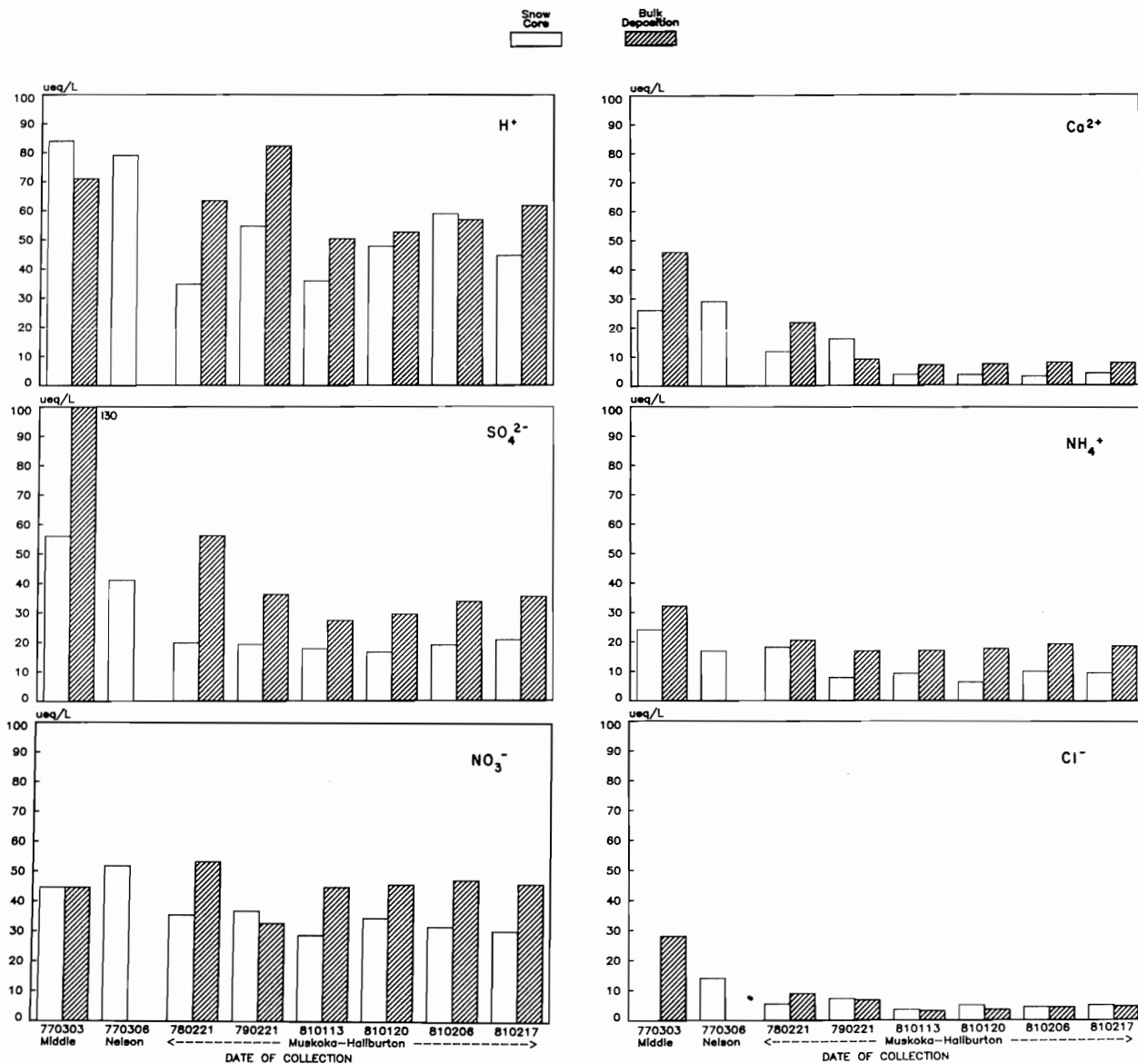


Figure 2. Histograms comparing the volume-weighted concentrations of H^+ , SO_4^{2-} , NO_3^- , Ca^{2+} , NH_4^+ , and Cl^- ($\mu\text{eq L}^{-1}$) in the snowpack (open bars) and corresponding cumulative bulk deposition (shaded bars) at various locations and times in central Ontario.

MUSKOKA-HALIBURTON SNOW PROFILE

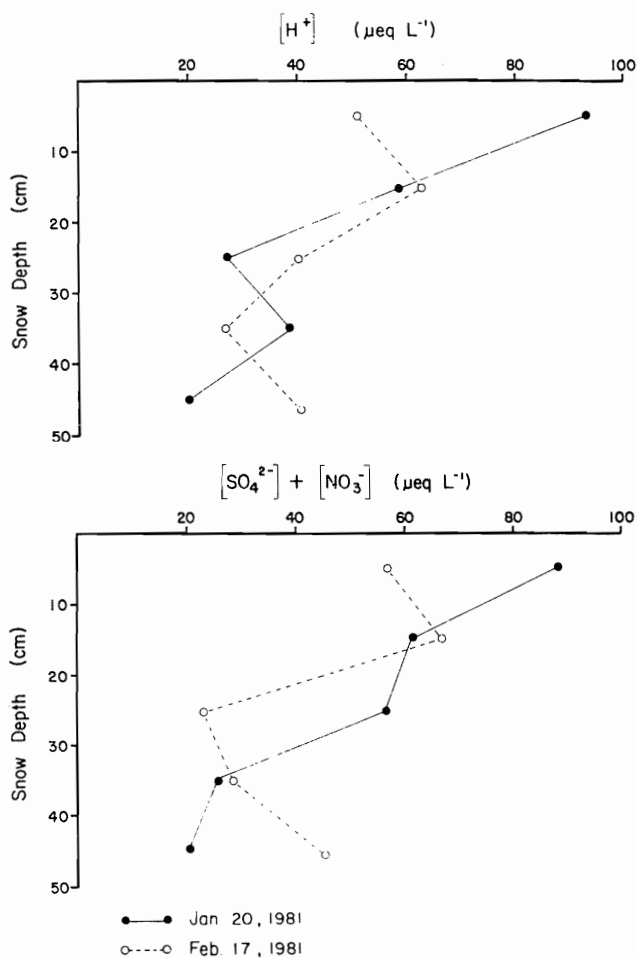


Figure 3. Variation in concentration of H^+ and $SO_4^{2-} + NO_3^-$ ($\mu\text{eq L}^{-1}$) with snow depth at Muskoka-Haliburton on January 20 and February 17, 1981.

showed an overall decrease with depth in protons and acid anions of $60-70 \mu\text{eq L}^{-1}$. The older layers of snow are reduced in concentration and since no similar trend in bulk deposition was recorded, this suggests that the loss occurs by migration of ions into the underlying soils.

The snow profile was re-sampled on Feb. 17 after the initiation of melting and the input of 38 mm of rain but before there was significant overall loss of the snowpack. Considerable change was noted in the distribution of H^+ and $SO_4^{2-} + NO_3^-$ within the profile. In particular, the near-surface snow showed reduced concentrations relative to the earlier profile, while elevated levels were observed at the bottom of the profile. It is impossible to determine whether these changes were due to melting or the input of rain but it once again is clear that the strong acid present in the snow has preferentially migrated downwards over the period between profile samplings.

Release of acid stored within the watershed (snowpack and soils) during the spring runoff resulted in major increases in H^+ concentration for the six streams

entering Harp Lake in Muskoka-Haliburton in 1979. Table 2 presents the pre- and post-melt pH values for the six streams as well as the minimum pH and the corresponding percent increase in H^+ relative to the pre-melt values.

Table 2. Minimum pH of six streams entering Harp Lake during the spring melt period of 1979. Pre- and post-melt pH values are provided as well as the increase in H^+ concentration at minimum pH relative to the pre-melt value.

Stream	pH			Increase ¹ [H^+] (%)
	prior to melt (ave. in Feb.)	minimum pH	post-melt (ave. in May)	
3	5.90	5.15	5.83	460
3A	5.85	5.49	6.29	130
4	6.25	5.42	6.33	580
5	5.47	4.89	5.37	280
6	5.72	5.34	5.80	140
6A	5.11	4.87	5.38	74

¹ Relative to Feb.

The pH of all the streams dropped below 5.5 during the runoff period, and in fact, two of the streams which have mean annual pH values < 5.5 (5 and 6A) had pH < 5.0. The increase in hydrogen ion concentration ranged from 74 to 580%. The pH depressions experienced by the Harp streams are comparable to or greater than those measured in 1978 (Jeffries *et al.* 1979). Many aquatic organisms pass through sensitive life-stages in the early spring (discussed in Harvey *et al.* 1981) and sudden reductions in pH such as those measured for the Harp Lake streams may be harmful. For example, Beamish (1976) reported reproductive failure for five fish species (including smallmouth bass, walleye and lake trout) in LaCloche Mountain lakes at pH values between 5.2 and 6.0.

The hydrographs and pattern of variation of H^+ , SO_4^{2-} , and NO_3^- concentrations for Harp inlet streams 3 and 4 from February to May 1979 are illustrated in Figures 4 to 6. Five of the six streams (including No. 3 and 4) exhibited peak H^+ concentrations coincident with peak discharge. This direct "flow- H^+ concentration" relationship suggests that H^+ export from the watershed is reaction rate controlled; that is, when water is quickly flushed through the system, insufficient time is available for the neutralization reactions to occur as they do before and after the runoff period. The relationship is unlike that reported for Norway (Johannessen *et al.* 1980) where peak H^+ concentrations preceded peak discharge and have been attributed to differential loss of H^+ from the snowpack during the first phases of snowmelt. Only one of the Harp streams (No. 6A) showed the "Norwegian style" of H^+ concentration discharge relationship. This stream also has the lowest annual mean pH of all the inlet streams. Given the snow core results discussed earlier, it is likely that differential release of H^+ also occurs at Muskoka-Haliburton. Hence, the general difference in H^+ pattern between the Harp Lake and the Norwegian basins is probably related to the greater thickness of overburden (1-3 m) at the former location, and therefore greater soil-meltwater interaction prior to discharge.

Sulphate and NO_3^- concentrations (Figures 5, 6) exhibited different patterns over the spring melt. Sulphate remained relatively constant over the entire period with some of the streams showing slightly lower concentrations after the melt period compared to before it. This observation is again different from that presented by Johannessen *et al.* (1980) who reported a strong, positive relationship between SO_4^{2-} and H^+ . Sulphate exchange reactions within the soil at Harp Lake are evidently fast enough to buffer the SO_4^{2-} concentration throughout spring melt.

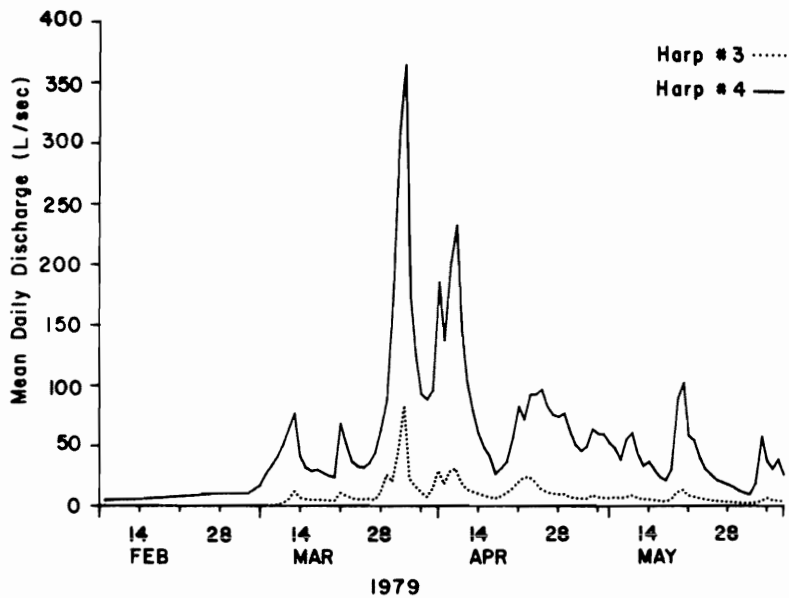


Figure 4. Hydrographs for Harp inlet streams 3 and 4 from February to May, 1979. Discharge is in L sec^{-1} .

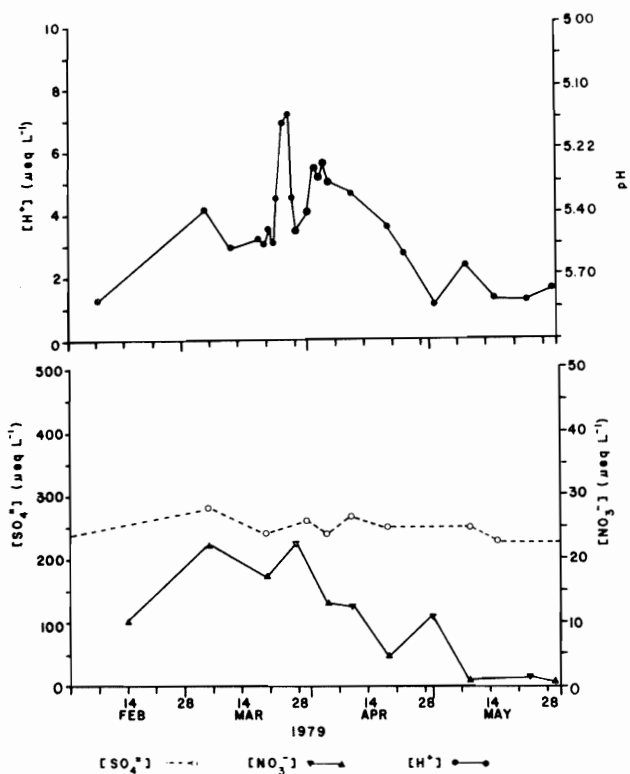


Figure 5. Harp inlet stream 3.

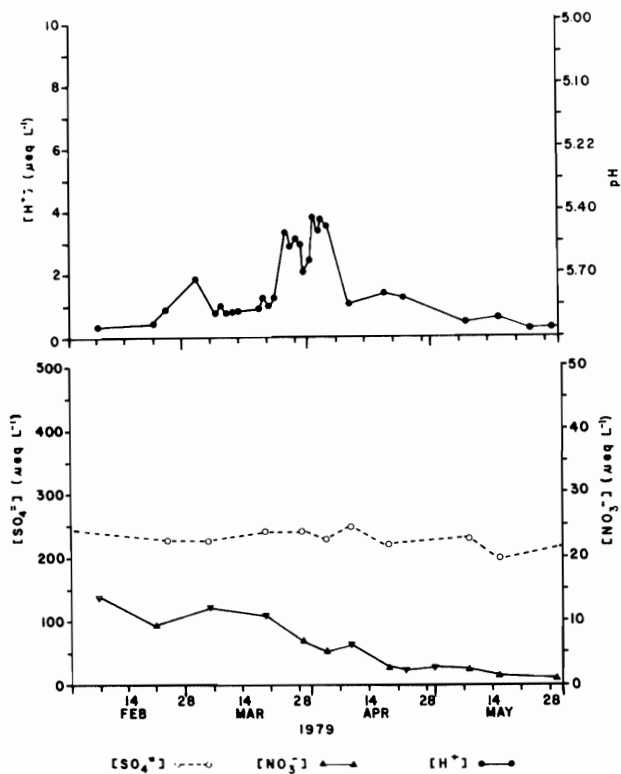


Figure 6. Harp inlet stream 4.

Pattern of variation in the concentrations of H^+ , SO_4^{2-} , and NO_3^- ($\mu\text{eq L}^{-1}$) throughout the spring melt period of 1979. Note the difference in magnitude of the SO_4^{2-} and NO_3^- scales.

Nitrate concentrations are generally elevated (compared to annual mean values) in the streams prior to and during the melt period and then drop to nearly undetectable levels in May, presumably due to biological utilization. At no time do NO_3^- concentrations approach those of SO_4^{2-} (note difference in scales in Figures 5 and 6) despite the fact that snowpack concentrations are nearly comparable.

Three of the six streams exhibited NO_3^- patterns similar to inlet 4 e.g., declining NO_3^- values associated with peak H^+ concentrations. These streams differ from those in the Adirondack region of New York State (Galloway *et al.* 1980) which exhibit coincident springtime NO_3^- and H^+ peaks. The remaining streams at Harp Lake showed variable NO_3^- to H^+ relationships (such as inlet 3, Fig. 5) where coincident peaks were observed at some times and not at others.

The $\text{SO}_4^{2-}:\text{NO}_3^-$ ratio (Table 3) is an indicator of the relative importance of the H_2SO_4 and HNO_3 inputs to the acidification process underway at Harp Lake. On a mass balance basis, all of the SO_4^{2-} input to the watersheds (in bulk deposition) is exported (Dillon *et al.* 1980) while a varying proportion of the NO_3^- input is retained depending on time of year. Annually >95% of the NO_3^- is retained at Harp Lake (Dillon *et al.* 1980). When the NO_3^- ion is retained in the watershed by biological utilization, there is an associated release of a basic anion (probably HCO_3^- or OH^- ; discussed in Harvey *et al.* 1981). Therefore in the situation where NO_3^- is retained, the HNO_3 input is not a net acidifying substance.

Table 3. Ratio of SO_4^{2-} to NO_3^- (both in $\mu\text{eq L}^{-1}$) in six streams entering Harp Lake during the spring melt period of 1979. The ratio for bulk deposition is also presented for comparison

Stream	[SO_4^{2-}]:[NO_3^-]		
	prior to melt (Jan. 24)	at minimum pH	post melt (May 30)
3	26	12	320
3A	10	10	220
4	17	40	250
5	25	38	670
6	15	14	670
6A	120	28	580

Bulk Deposition - Annual Ratio = 2.1 (Aug. 1976 - July 1978; Scheider *et al.* 1979a)
 Winter Ratio = 1.3 (Nov. 1978 - Feb. 1979; Jeffries, unpub. data)

The $\text{SO}_4^{2-}:\text{NO}_3^-$ ratios for the streams are always very much greater than that for bulk deposition (Table 3), and values in May (and throughout the summer and autumn) are very large due to utilization of NO_3^- . At the time of minimum pH in the spring of 1979, four of the six inlet streams exhibited constant or increased $\text{SO}_4^{2-}:\text{NO}_3^-$ ratios relative to pre-melt values. Sulphuric acid is clearly the dominating acidifying strong acid in these cases. Two of the streams showed reduced ratios indicating an increased significance for HNO_3 although those two values are still at least 10-fold greater than the winter ratio for bulk deposition from November 1978 to February 1979.

SUMMARY

1. The coefficient of variation in snow core concentrations of H^+ , SO_4^{2-} , NO_3^- , NH_4^+ , Ca^{2+} , Cl^- and Al at urban and rural sites in central Ontario ranged from 5.6 to 137%

and was generally < 45%. This degree of variation is small compared to the temporal variation in concentration of precipitation samples. Greater variation was evident at the urban site (Middle Lake) due to the influence of roads.

2. Higher snow core concentrations of H^+ , SO_4^{2-} , NO_3^- , Ca^{2+} , and Cl^- at Sudbury compared to Muskoka-Haliburton reflect the greater dry deposition of SO_2 and urban influences at the former location.
3. Snow core concentrations were generally less than corresponding bulk deposition concentrations (except Cl^-). The degree of difference between snow core and bulk concentrations is variable; SO_4^{2-} is notably more depleted from the snowpack than H^+ or NO_3^- .
4. Sampling of the snowpack in profile showed that H^+ and $SO_4^{2-} + NO_3^-$ concentrations generally decreased with depth and were presumably lost from the snowpack by migration into the underlying soils. This can occur without significant melting.
5. During the snowmelt period of 1979, all six streams entering Harp Lake in Muskoka-Haliburton showed pH depressions. Increase in H^+ concentrations ranged from 74 to 580% relative to pre-melt values.
6. Peak H^+ concentrations were observed to coincide with peak discharge for five of the six inlet streams. This is a different situation than in Norway where peak H^+ precedes peak discharge (Johannessen *et al.* 1980). Similarly the near constant SO_4^{2-} concentrations observed throughout the melt period are different from Norwegian observations where SO_4^{2-} varies in the same manner as H^+ .
7. Nitrate concentrations are greater during pre-melt and melt periods than during post-melt periods when values are extremely low due to biological uptake. For three of the six streams, declining NO_3^- concentrations coincided with peak H^+ . This situation is different than that observed in Adirondack streams where peak H^+ and NO_3^- coincide (Galloway *et al.* 1980). The remaining three inlet streams exhibited variable NO_3^- to H^+ relationships.
8. Consideration of the ratio of strong acid anions in the stream waters (e.g., $SO_4^{2-}:NO_3^-$) suggests that sulphuric acid is the dominant acidifying strong acid at Muskoka-Haliburton.

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