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The role of ammonium and nitrate retention in the acidification of lakes and forested catchments

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Abstract. The relative contribution of HNO_3 to precipitation acidity in eastern Canada has increased in recent years leading to some concern that the relative importance of NO_3^- deposition in acidification of terrestrial and aquatic ecosystems may increase. To gauge the extent of this impact, annual mass balances for NO_3^- and NH_4^+ were calculated for several forested catchments and lakes in Ontario. Retention of NH_4^+ (R_{NH_4}) by forested catchments was consistently high compared to retention of NO_3^- (R_{NO_3}) which was highly variable. Retention of inorganic nitrogen was influenced by catchment grade and areal water discharge. In lakes, the reciprocals of retention of NO_3^- and NH_4^+ were linearly related to the ratio of lake mean depth to water residence time ($\bar{z}/\tau$; equal to areal water discharge), and retention did not appear to be a function of degree of acidification of the lakes. Net N consumption-based acidification of lakes, defined as the ratio of annual NH_4^+ mass to NO_3^- mass consumption, was negatively correlated with $\bar{z}/\tau$ and N consumption-related acidification was most likely to occur when $\bar{z}/\tau$ was $< 1.5\,\mathrm{m\,yr^{-1}}$.

If retention mechanisms are unaffected by changes in deposition, changes in deposition will still result in changes in surface water concentrations although the changes will be of similar proportions. Therefore, 'NO_3^- saturation' should not be defined by concentrations alone, but should be defined as decreasing long-term, average NO_3^- retention in streams and lakes in response to long-term increases in NO_3^- deposition. Analysis of survey data will be facilitated by grouping lakes and catchments according to similar characteristics.

Introduction

Nitrogen oxide emissions in eastern North America and nitrate deposition and concentration in precipitation in central Ontario have not changed significantly in the past decade (Dillon et al. 1988). In contrast, SO_x emissions, deposition and concentrations have decreased, resulting in a gradual increase in the equivalent ratio of NO_3^- to SO_4^{2-} in deposition in central Ontario from 0.43 in 1976/77 to 0.68 in 1985/86 (Dillon et al. 1988). The maximum possible contribution of NO_3^- to precipitation acidity (NO_3^-/H^+) has also increased in recent years whereas the maximum contribution of SO_4^{2-} has not changed (Dillon et al. 1988). These trends in deposition had led to concern that the relative importance of NO_3^- deposition to acidification of terrestrial and aquatic ecosystems may increase (McLean 1981; Galloway & Dillon 1983; Aber et al. 1989). Concern has also been expressed about the acidification effects of N deposition in Europe (Grennfelt & Hultberg 1986).

The contribution of both NO_3^- and NH_4^+ deposition to long-term acidification is controlled by biological consumption and production of each, which generates or consumes alkalinity, respectively (Brewer & Goldman 1976; Schindler et al. 1985; Grennfelt & Hultberg 1986). Since NH_4^+ is generally a preferred source of N for assimilation (Syrett 1981), high NH_4^+ deposition rates may generate considerable acidity via high NH_4^+ assimilation and consequent inhibition of NO_3^- assimilation rates. High NH_4^+ deposition rates in some parts of Europe are considered to be a major cause of acidification of ecosystems (van Breemen et al. 1982). As a further complication, microbiological processes such as mineralization, denitrification and nitrification may be affected by acidification and high SO_4^{2-} concentrations (Kowalenko 1979; Strayer et al. 1981; Knowles 1982; Klein et al. 1983; Rudd et al. 1988). For example, Rudd et al. (1988) demonstrated that acidification to a pH of 5.4–5.7 resulted in the cessation of nitrification in small soft-water lakes in Canada.

Mass balance studies show that there is a wide range of values for retention of NO_3^- by lakes and catchments (Henriksen & Wright 1977; Likens et al. 1977; Galloway et al. 1980, 1983; Wright & Johannessen 1980; Kerekes et al. 1982; Wright 1983; Hemond & Eshleman 1984; Kelly et al. 1987). Sulphate retention by lakes and catchments is usually much lower than NO_3^- retention, indicating less chemical and biological reactivity (Schindler et al. 1976, 1986; Galloway et al. 1980; Wright & Johannessen 1980; Hemond & Eshleman 1984; Kelly et al. 1987). Processes resulting in retention of NO_3^- within a lake or catchment are considered to generate an equivalent or near-equivalent amount of alkalinity, while those resulting in NH_4^+ retention generate an equivalent amount of mineral acidity.

Because a number of biological processes result in the production or consumption of either NH_4^+ and NO_3^-, including nitrification, denitrification, decomposition of organic matter, and, indirectly, nitrogen fixation, mass balances for lakes and/or catchments based on only the input and output of the two chemical species cannot adequately describe the inorganic nitrogen cycle in these systems. However, the role of NH_4^+ and NO_3^- deposition in acidifying these systems can be evaluated by measuring NH_4^+ and NO_3^- input/output budgets. Subsequent comparison of NH_4^+ and NO_3^- input/output budgets between systems must then take into account differences between catchment/lake characteristics which may affect consumption such as forest age and type, precipitation, runoff, gradient, flushing rate, etc. before evaluating the effects of deposition.

In this paper annual NO_3^- and NH_4^+ mass balances for seven subcatchments, two lakes, and two combined lake-catchment systems in central Ontario are reported for eight years (1976–1984). The results are compared to those from several lakes and forested stream catchments experiencing a range of N deposition rates in different regions of Canada, in the USA and in Scandinavia. We attempt to distinguish several factors which affect ability of lakes and forested catchments to retain NO_3^- and NH_4^+ in order to determine whether retention is affected by high deposition rates and/or catchment/lake differences.

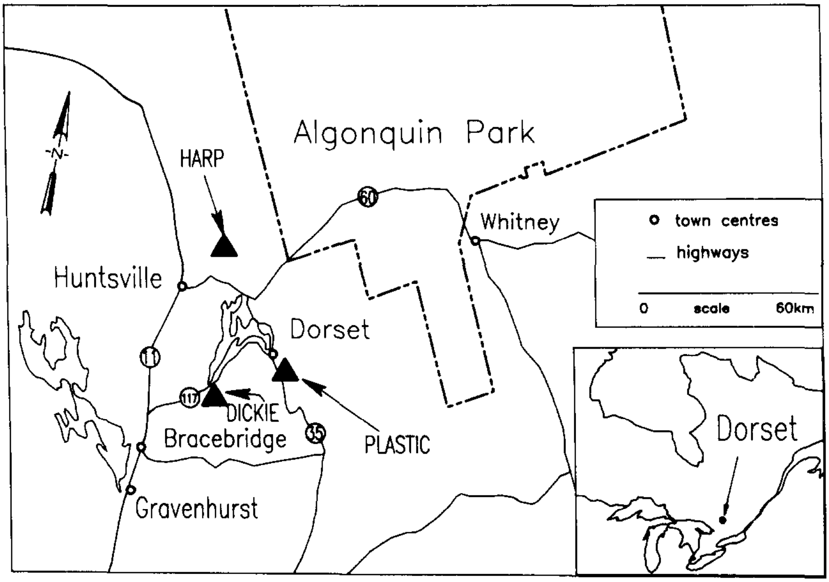

Fig. 1. Location of the study lakes and their subcatchments.

Study area

Harp and Plastic Lakes are located in the Muskoka–Haliburton area of central Ontario (Fig. 1). Plastic Lake is the more acidic (mean volume-weighted alkalinity 15 μeq/L in 1980/81) with measurable decreases in pH and alkalinity occurring since 1979 (Dillon et al. 1987). Harp Lake is less acidic but is still considered acid-sensitive with mean volume-weighted alkalinity of 61 μeq/L and pH 6.3. The lakes are phosphorus-limited and oligotrophic with mean annual total phosphorus ranging from 6.7–8.2 $\mu g\,P \cdot L^{-1}$ and the total N/total phosphorus mass ratios ranging from 33–48 in Harp Lake during the period 1976–1988. Total phosphorus ranged from 4.6–7.8 $\mu g\,P \cdot L^{-1}$ and the total N/total phosphorus ratio ranged from 35–44 in Plastic Lake during the period 1979–1988.

Catchment characteristics are listed in Table 1. The catchments are underlain by Precambrian metamorphic silicate bedrock. The Plastic Lake catchment has surficial deposits of thin Pleistocene glacial till (< 1 m) and small areas of peat overlying layers of sand. The Harp Lake catchment has extensive till plains (> 1 m) with some areas of thin till and exposed rock ridges. Smaller deposits of sand and peat (overlying sand) occur. More detailed descriptions of the geology and physiography of the catchments can be found in Jeffries & Snyder (1983), LaZerte & Dillon (1984), Girard et al. (1985), Seip et al. (1985) and Dillon et al. (1987).

The catchments are primarily forested (predominately coniferous at Plastic, deciduous at Harp) with some cottage development around Harp Lake. The Plastic Lake catchment has not been logged in at least 70 years, while selective logging in parts of the Harp Lake catchment has been conducted periodically,

Table 1. Catchment characteristics of study sites in the Muskoka–Haliburton area of central Ontario.

	Area (ha)	Bedrock geology[a]	Surficial geology[b]
Harp Lake, Ontario			
Inlet 3	26.0	1	8
3A	19.7	1, 3	8
4	119.1	3	8, 6, 7
5	190.5	1	8, 6, 5, 7
6	10.0	2, 1	6, 8
6A	15.3	1	6
Ungauged	90.1	1, 3	8, 6
Lake	71.4	–	–
Outflow	542.0[c]	–	–
Plastic Lake, Ontario			
Inlet 1	23.3	4	6
Ungauged	69.2	4	6
Lake	32.1	–	–
Outflow	124.6[c]	–	–

[a] 1. Granitized biotite and horneblende gneiss; 2. Diorite; 3. Amphibolite and schist; 4. Orthogneiss.
[b] 5. Peat over sand; 6. Thin till and rock ridges; 7. Sand; 8. Minor till plain.
[c] Includes lake area and all drainage into the lake.

the last time being 30–40 years ago. Harp Lake has six major tributaries which each drain between 10 and 191 ha of the catchment (Table 1). Plastic Lake has one major inflow and several ephemeral inflows; only the former was treated as a separate subcatchment in this study.

Methods

The input of NO_3^- and NH_4^+ to the subcatchments from the atmosphere was estimated by measuring bulk deposition rates; methods are described in detail in Dillon et al. (1988). Although bulk deposition appears to measure wet and particulate inputs adequately, it may not effectively include inputs of gaseous precursors of NO_3^- (NO_x), resulting in an underestimate in the deposition of NO_3^-.

The output of NO_3^- and NH_4^+ from each of the subcatchments for eight hydrologic years (June 1, 1976–May 31, 1984) was estimated by combining continuous measurements of stream discharge with instantaneous estimates of substance concentration. Weirs or flumes were installed at the mouth of the seven major inlet streams as well as at the two lake outflows. Discharge was calculated by applying a measured water level-discharge relationship to the continuously-measured stream level (Scheider et al. 1983). Water samples were collected approximately weekly, except during periods of high flow (especially snowmelt), when more samples were taken, and in the winter, when sampling frequency was reduced. Most streams were sampled 35–50 times in any of the 8 years, although the number varied from 28–132. Sample collection techniques were described in detail in Locke and Scott (1986), while analytical methods are outlined in Ontario Ministry of the Environment (1983). Total inorganic nitrogen (TIN) was calculated as NO_3^- plus NH_4^+.

For catchment budgets, precipitation was considered to be the only input. Outputs were expressed per m^2 of catchment.

The NO_3^- and NH_4^+ outputs from the lakes were measured in an identical manner. The total inputs to each of the lakes were calculated as the sum of the input(s) from the subcatchments, the input from the ungauged portion of the catchment (prorated on an areal basis) and the atmospheric deposition directly on the lakes' surfaces. Retention in the lake was calculated as the difference between total inuts and output divided by total input.

Budgets for the combined lake-catchment systems were calculated from atmospheric deposition to the total land and water area and output via the lakes' outflows. Only inputs and outputs of lakes of NH_4^+ and NO_3^- were considered. Changes in internal N storage were not considered.

Precipitation samples were obtained with bulk collectors in Ontario and British Columbia and with wet-only event samplers at Lac Laflamme, Quebec and Kejimkujik Lake, Nova Scotia. The Kejimkujik data were presented as estimated wet + dry deposition by Kerekes et al. (1982) but only wet deposition is used here.

The extent of spurious self-correlation was investigated by correlation analysis of a randomly generated data set consisting of 100 lake-years of observations of N concentrations in an inflow stream, the lake outflow and precipitation, discharge from the inflow stream and outflow, precipitation depth, lake surface area and mean depth. The simulated variables were normally distributed using realistic scales.

Results and discussion

Catchment retention

The retentions of NO_3^- (R_{NO_3}), NH_4^+ (R_{NH_4}) and TIN (R_{TIN}) in the Harp and Plastic Lake subcatchments, lakes and lake/catchment systems are shown in Table 2. There was, with few exceptions, very little difference between years in retention of any of the three parameters as evidenced by the small coefficients of variation (< 10% in most cases).

A minimum of 94% (± 3%) of the NH_4^+ deposited on the subcatchments was retained. The retention of NO_3^- was lower (0.52–0.98) and more variable, although for 37 of 53 catchment-years, it was > 0.8. The catchment with the lowest average retention (0.52 ± 0.35 in Harp 3A) was affected by a single year (1977/78) when R_{NO_3} was −0.05 (precipitation input < export). In contrast,

Table 2. Retention of NH_4^+, NO_3^- and TIN (= NO_3^- + NH_4^+) in study lakes and catchments. Means ± 1 SD are shown. For Harp Lake, n = 8 yr (1976–84); for Plastic, n = 5 (1979–84).

Location	R_{NO_3}	R_{NH_4}	R_{TIN}
Harp Lake, Ontario			
Inlet 3	0.79 ± 0.13	0.97 ± 0.02	0.86 ± 0.08
3A	0.52 ± 0.35	0.99 ± 0.01	0.70 ± 0.20
4	0.91 ± 0.03	0.94 ± 0.03	0.92 ± 0.02
5	0.87 ± 0.07	0.95 ± 0.02	0.90 ± 0.04
6	0.75 ± 0.08	0.96 ± 0.02	0.84 ± 0.05
6A	0.98 ± 0.01	0.99 ± 0.01	0.98 ± 0.01
Lake	0.57 ± 0.13	0.88 ± 0.06	0.67 ± 0.09
Catchment + Lake	0.79 ± 0.29	0.98 ± 0.01	0.92 ± 0.02
Plastic Lake, Ontario			
Inlet 1	0.97 ± 0.01	0.99 ± 0.01	0.98 ± 0.07
Lake	0.81 ± 0.02	0.79 ± 0.10	0.80 ± 0.04
Catchment + Lake	0.85 ± 0.01	0.95 ± 0.03	0.85 ± 0.01

R_{NO_3} was 0.93 in 1976/77. Excluding the low value, the mean for Harp 3A was 0.60 ± 0.29, which was still the lowest average for any subcatchment. The retention of TIN ranged from 0.70 (±0.20) to 0.98 (±0.07), with the lowest value (Harp 3A) again influenced by the one year's results in 1977/78.

When each lake and its catchment are considered as a whole, both were extremely effective NH_4^+ sinks (0.98 and 0.95 for Harp and Plastic, respectively). The Plastic system was equally effective in retaining NO_3^- (R_{NO_3} = 0.95), while the Harp system was less so (R_{NO_3} = 0.79).

Measured NH_4^+ retention for stream catchments and lake/catchment systems in Europe and North America including Harp and Plastic was high ranging from 0.81–0.99 for loadings up to 40 meq m^{-2} yr^{-1}, the highest deposition rate in North America (Barrie and Hales 1984). The exception was Jamieson Creek in British Columbia with R_{NH_4} = 0.05 (Table 3).

Mean annual R_{NO_3} for stream catchments reported in Tables 2–4 was variable, ranging from 0.15 for Hubbard Brook, New Hampshire to 0.98 in Harp 6A for loadings up to 83 meq m^{-2} yr^{-1}. R_{NO_3} was < 0 at Hubbard Brook during 3 of the 11 years (Likens et al. 1977) which indicates conditions promoting nitrification, or that dry (unmeasured) deposition was high. This high between-year variability at Hubbard Brook was similar to that at the Harp 3A catchment. Relatively low R_{NO_3} occurred in the Lac Laflamme, Batchawana South and Turkey Lakes catchments and certain Harp Lake subcatchments during some years. Batchawana South and Turkey Lakes have short residence times and rapidly flushing lakes upstream which probably resulted in the low total catchment R_{NO_3} as discussed in the next section. Lohi Lake also has a short residence time and an upstream lake. The upstream lake, Clearwater, has a low R_{NO_3} of 0.51.

R_{NO_3} was also variable in catchment/lake systems (Tables 2—4), ranging from

Table 3. Retention of NH_4^+, NO_3^- and TIN (= NO_3^- + NH_4^+) in Canadian catchments and lake/catchment systems. Numbers are mean ± 1 SD; *n* is number of years of data.

Location	*n*	R_{NO_3}	R_{NH_4}	R_{TIN}	References
Lake 223, Ontario					
Catchment + Lake	8 (1976–83)	0.98 ± 0.02	0.99 ± 0.01	0.99 ± 0.02	Schindler et al. (1976, 1980a, 1980b)
Lake 239, Ontario					
Northwest Subcatchment	13 (1971–83)	0.94 ± 0.11	0.98 ± 0.01	0.96 ± 0.05	Newbury et al. (1980)
Catchment + Lake		0.95 ± 0.04	0.98 ± 0.01	0.98 ± 0.02	Linsey et al. (1987)
Batchawana South, Ontario					
Mean of 4 Subcatchments	2 (1981–83)	0.63	0.97	0.77	Jeffries & Semkin (1982, 1983)
		0.72	0.91	0.80	Semkin & Jeffries (1983)
Catchment + Lake					
Turkey Lake, Ontario					
Mean of 3 Subcatchments	2 (1981–83)	0.40	0.97	0.63	Jeffries (unpub. studies)
		0.63	0.94	0.78	
Catchment + Lake					
Turkey Lakes, Ontario					
Subcatchment 31	1 (1983)	0.37	0.89	0.57	
Kejimkujik Lake, Nova Scotia					
Catchment + Lake	1 (1979–80)	0.99	0.81	0.94	Kerekes et al. (1982)
Lac Laflamme, Quebec					
Catchment + Lake	4 (1981–84)	0.64	-	-	Papineau (1987)
Lohi Lake, Ontario					
Catchment + Lake	2 (1977–79)	0.91	0.94	0.92	Jeffries et al. (1984)
Clearwater Lake, Ontario					
Subcatchment 1	2 (1977–79)	0.97	0.96	0.97	Scheider (1984)
Subcatchment 2		0.96	0.97	0.97	Yan & Dillon (1984)
Subcatchment 4		0.95	0.87	0.92	
Catchment + Lake		0.90	0.94	0.92	
Jamieson Creek, BC					
Catchment	1 (1970–71)	0.27	0.05	0.13	Zeman (1975)

Table 4. Reported mean annual retention, precipitation inputs and outputs for NH_4^+ and NO_3^- in Scandinavian and northeastern United States catchments and lake/catchment systems.

System	Years	Input meq/m²/yr	Output meq/m²/yr	*R*	References
NO_3^-					
Bickford Watershed	1981–83				
West Wachusett Brook		29.5	1.0	0.97	Hemond & Eshleman (1984)
Provencial Brook		29.5	0.79	0.97	
Bickford Reservoir		82.5	58.2	0.29	
Hubbard Brook	1963–74	30.7	26.0	0.15	Likens et al. (1977)
Adirondacks	1978–80				
Panther[2]		–	–	0.35	Galloway et al. (1980)
Sagamore[2]		–	–	0.43	
Woods[2]		–	–	0.54	
Birkenes	1972–78	53	8	0.85	Wright & Johannesson (1980)
Langtjern	1974–80				
Subcatchment 2		25.3	1.1	0.96	Henriksen & Wright (1977)
Subcatchment 3		25.3	0.9	0.96	
Catchment + Lake		25.3	1.4	0.94	
NH_4^+					
Hubbard Brook	1963–74	16.0	1.9	0.88	Likens et al. (1977)
Adirondacks	1978–80				
Panther[2]		–	–	0.92	Galloway et al. (1980)
Sagamore[2]		–	–	0.98	
Woods[2]		–	–	0.87	
Langtjern[2]	1973–75	22.5	–	–	Henriksen & Wright (1977)

[1] Dry deposition estimated using Cl method.
[2] Catchment + lake.

0.35 in the Panther catchment/lake system in the Adirondacks (the least sensitive of the three Adirondack sites), to 0.99 in the Kejimkujik catchment/lake system in Nova Scotia (which is highly acidified). The three Adirondack systems span a range from very acidic to neutral yet NO_3^- retention was low in all three. Clearly, acidification has little effect on R_{NO_3}.

Grennfelt & Hultberg (1986) reported that R_{NO_3} was < 0 in several forested catchments in Denmark, Belgium and Czechoslovakia when both high NH_4^+ (54–90 meq m^{-2} yr^{-1}) and high NO_3^- deposition (39–79 meq m^{-2} yr^{-1}) occurred. R_{NH_4} was high over the entire range of loadings (2.5–106 meq m^{-2} yr^{-1}). At the highest NH_4^+ deposition rate, however, a relatively high R_{NO_3} of 0.79 occurred suggesting that denitrification was unaffected (assuming that high NH_4^+ deposition inhibits NO_3^- assimilation).

NH_4^+ adsorption by soils is typically high whereas NO_3^- is mobile and in comparison is readily leached (Szperlinski and Badowska 1977a, 1977b). NH_4^+ exported from large agricultural catchments in southern Ontario was highly correlated with suspended solids, unlike NO_3^- (Coote et al. 1982). NO_3^- mobility implies that runoff exerts some control over export and, hence, retention.

Another variable which may affect R_{NO_3} is catchment grade which is consistent with the behaviour of NO_3^- as a mobile anion. The correlation coefficient between mean NO_3^- export (n = 8 years) and average grade was +0.66 for 32 small stream catchments in central Ontario (P. Dillon, unpub. data). Steep grade may be related to the low retention observed in the Jamieson Creek and Hubbard Brook catchments and the Adirondack catchment/lake systems.

It appears that NO_3^- is typically the dominant inorganic N species exported not only from the Harp and Plastic catchments but also from many other catchments, regardless of N source, because of high NH_4^+ retention and microbial transformations. Klein et al. (1983) reported high rates of NO_3^- formation in acid forest soils. Although autotrophic nitrification is inhibited by low pH, heterotrophic nitrification is not (Lang & Jagnow 1986). It follows, therefore, that NO_3^- exported from forested catchments is not necessarily derived from atmospheric NO_3^-.

The retention of total inorganic N, R_{TIN}, differed between adjacent Harp stream catchments, although TIN deposition was identical; furthermore, R_{TIN} did not appear to be a function of wet deposition over the range of 60–80 meq m^{-2} yr^{-1} in this data set (Fig. 2). The occurrence of high wet deposition in the Dorset area in 1977/78 and 1978/79 was not due to higher precipitation (Dillon et al. 1988). R_{NO_3} showed no relationship to wet deposition using data from Ontario, British Columbia, Hubbard Brook and Norway (Fig. 3). However, there was a weak relationship between R_{NO_3} and water discharge (Fig. 4) which is also consistent with the behaviour of NO_3^- as a mobile anion. Clearly, NO_3^- retention is affected by catchment characteristics such as grade and runoff and not solely by NO_3^- deposition. It has also been argued that vegetation type and age affect retention (Hemond & Eshleman 1984). Therefore, searching for evidence of NO_3^- saturation in a survey of forested catchments receiving a range of NO_3^- deposition rates will be misleading unless relevant catchment characteristics are taken into account. Significant decreases in annual retention within

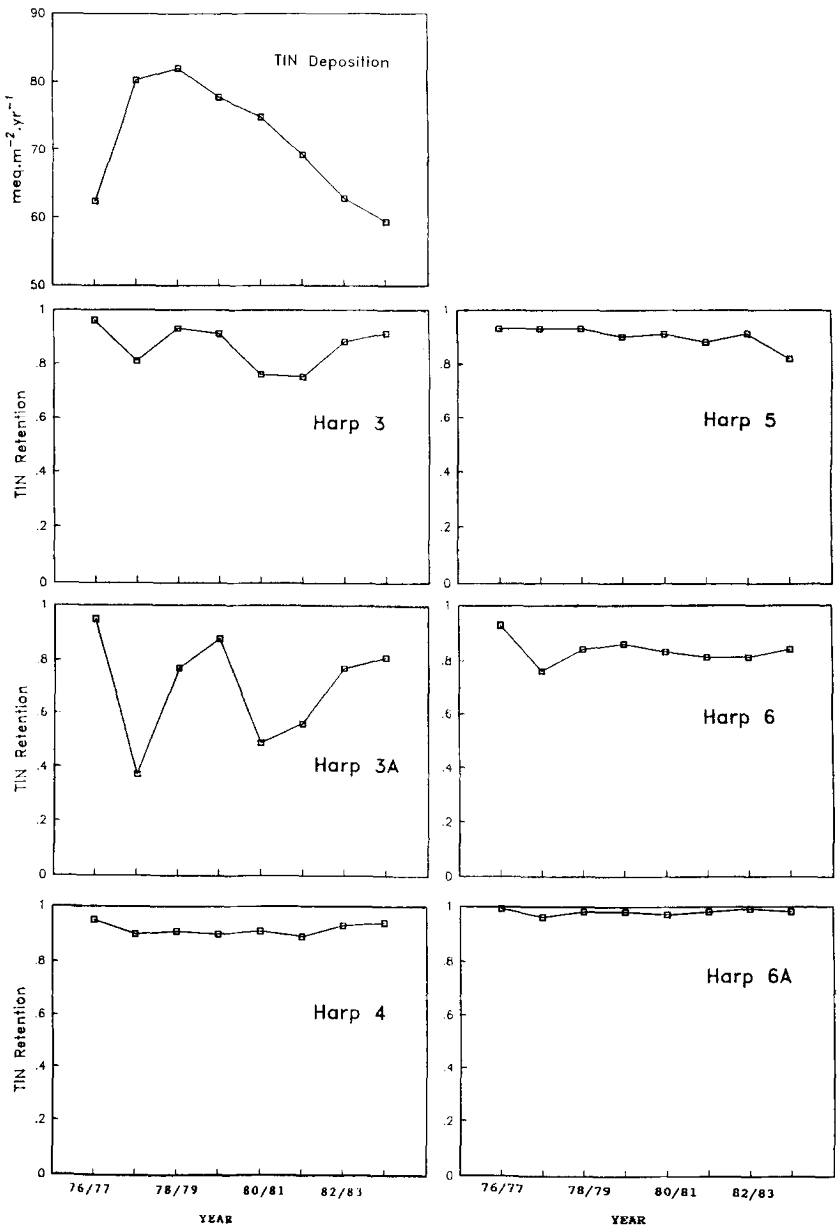

Fig. 2. Annual wet deposition and TIN retention (R_{TIN}) for Harp stream catchments.

a catchment in conjunction with significant increases in deposition would constitute evidence of NO_3^- saturation regardless of changes in internal N storage pools. However, given the inherent variability in some systems such as Harp 3A and Hubbard Brook, long-term data sets will be necessary to factor out annual variation.

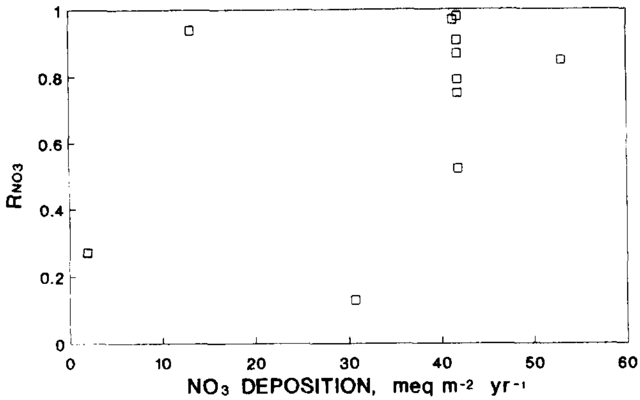

Fig. 3. Annual stream catchment R_{NO_3} vs. NO_3^- deposition for Harp (3, 3A, 4, 5, 6, 6A), Lake 223 (Northwest), Hubbard Brook, Birkenes, and Jamieson Creek, British Columbia.

Lake retention

Retention of both nitrogen species was lower in Plastic and Harp Lakes than in the subcatchments of the lakes; i.e. the lakes were less efficient NO_3^- and NH_4^+ sinks, although the residence time of the water in the lakes was almost certainly much greater than it was in any of the catchments (Table 2).

The NO_3^- retention model of Kelly et al. (1987), which is based on the Vollenweider (1969) model relates lake retention, R_{NO_3}, to lake mean depth, $\bar{z}$, and water residence time, τ:

$$R_{NO_3} = \frac{S_{NO_3}}{\bar{z}/\tau + S_{NO_3}} \tag{1}$$

where S_{NO_3} is a net mass transfer coefficient ($m\,yr^{-1}$) and is considered to be a

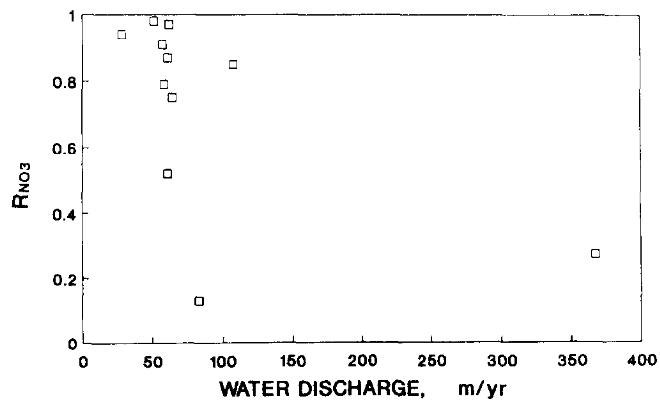

Fig. 4. Annual stream catchment R_{NO_3} vs. water discharge for Harp (3, 3A, 4, 5, 6, 6A), Lake 223 (Northwest), Hubbard Brook, Birkenes and Jamieson Creek, British Columbia.

Table 5. Mean Lake NO_3^-, NH_4^+ and TIN retention, water residence time (τ, in yr), mean depth ($\bar{z}$ in m), pH, and mass transfer coefficients (S, in $m\,yr^{-1}$) in several Ontario lakes.

Lake	Budget years	R_{NH_4}	R_{NO_3}	R_{TIN}	τ	$\bar{z}$	S_{NO_3}	S_{NH_4}	S_{TIN}	pH
239	1976–83	0.89	0.73	0.82	6.2	10.9	4.76	14.67	7.97	6.5–6.8
223	1976–83	0.84	0.98[2]	–	8.7	7.2	25	6.92	–	5.0–6.7
Harp	1976–84	0.88	0.57	0.67	2.5	13.3	6.50	48.36	10.80	6.2–6.3
Plastic	1979–84	0.79	0.81	0.80	3.0	8.0	11.38	9.89	10.68	5.7–5.9
Lohi	1977–79	0.51	0.45	0.46	1.0	6.2	5.07	6.33	5.28	4.7–5.3
Clearwater	1977–79	0.74	0.51	0.61	3.7	8.3	2.33	3.03	3.50	4.1–4.5
Batchawana South	1981–83	0.29	0.22	0.24	0.3	3.3	3.10	4.49	3.47	5.9
Turkey	1981–83	0.34	0.28	0.29	0.9	12.2	5.27	6.81	5.54	6.7
Regression estimate using Eq. 2 (±95% confidence limits), n = 8 lakes								4.15 ±3.65	5.59 ±4.84	4.31 ±4.50
Crystal[1]	1984	–	0.99	–	25.	10.6	42.0	–	–	–
$302N$[2]	1982–85	–	0.69	–	5.8	5.7	4.7	–	–	6.2–6.6
$302S$[2]	1982–85	–	0.89	–	8.3	5.1	6.0	–	–	5.6–6.6
Langtjern[1]	1972–78	–	0.36	–	0.20	2.4	6.8	–	–	4.6–4.8
Regression estimate using Eq. 2 (±95% confidence limits), n = 12 lakes							4.84 ±2.63	–	–	

[1] Data from Kelly et al. (1987)
[2] Data from C.A. Kelly (pers. comm.)

constant if denitrification is the major NO_3^- removal mechanism (Kelly et al. 1990). Note that $\bar{z}/\tau$ ($m\,yr^{-1}$) is equivalent to the areal water discharge from the lake.

Equation 1 can be transformed into a linear relationship:

$$\frac{1}{R_{NO_3}} = \frac{1}{S_{NO_3}} \cdot \frac{\bar{z}}{\tau} + 1. \tag{2}$$

The data presented in Table 5 are highly correlated when linearly transformed ($r = 0.89$) and are plotted in Fig. 5a. This suggests that the retention model is a reasonable description of in-lake processes. S_{NO_3}, calculated as the reciprocal of the slope (±95% confidence limits) was $4.8 \pm 2.6\,m\,yr^{-1}$. Agreement is good except for Batchawana South and Dart's Lakes which have very short and probably highly variable τ (Table 5). Hence, $\bar{z}/\tau$ appears to strongly control R_{NO_3}, at least within the range of NO_3^- loadings (9–321 $meq\,m^{-2}$ lake surface yr^{-1} for the Ontario lakes) and lake pH values observed.

Since R_{NO_3} increases with decreasing $\bar{z}/\tau$, smaller retention by lakes compared to stream catchments (Tables 2–5) reflects less contact between NO_3^- and microbial communities primarily responsible for consumption in these lakes (Keeney et al. 1971; Knowles & Lean 1987). Variability of S_{NO_3} (Tables 2 and

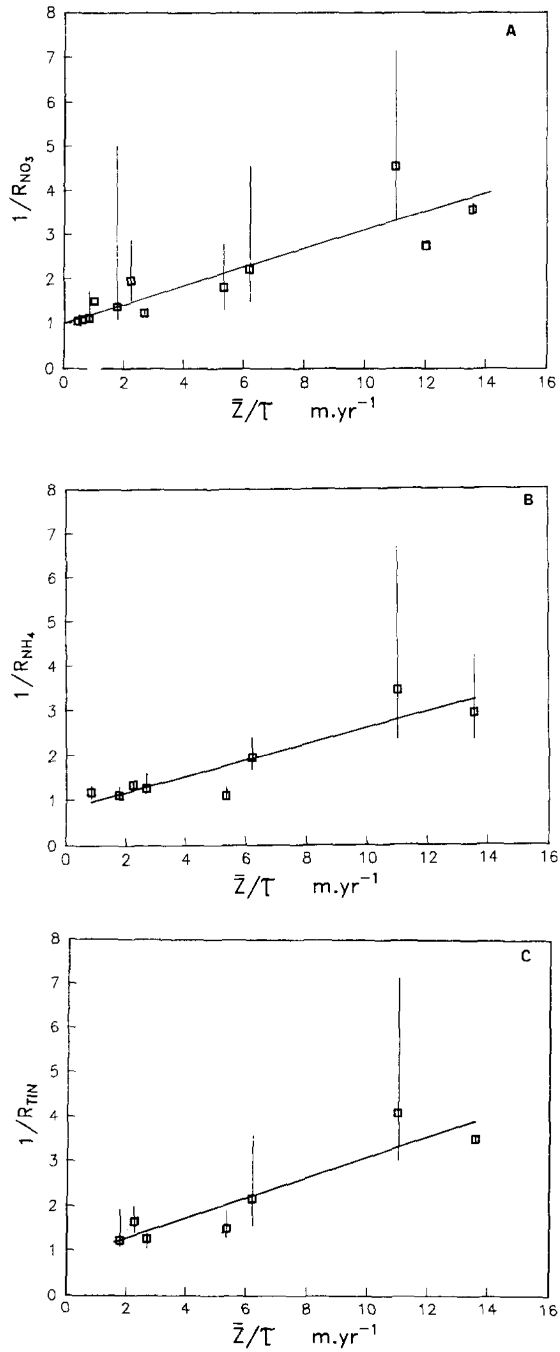

Fig. 5. Reciprocal of mean lake NO_3^-, NH_4^+ and TIN retention vs. $\bar{z}/\tau$ using data from Table 5. The line was fit with ordinary least squares regression. Error bars are ranges.

5) may also reflect *in situ* variability in turbulence, and microbial denitrification, nitrification and assimilation rates.

Kelly et al. (1990) offer convincing evidence that S_{NO_3} is also a function of the type of biological process responsible for NO_3^- consumption. S_{NO_3} values are $< 11\ m\,yr^{-1}$ when denitrification is the primary NO_3^- sink and $> 25\ m\,yr^{-1}$ when algal consumption is the primary sink. Furthermore, they argue that lakes with high S_{NO_3} values are capable of very efficient NO_3^- consumption. Therefore, large S_{NO_3} decreases in a lake in which algae dominate NO_3^- consumption can be taken as evidence that N saturation of the algal community is occurring. However, in some lakes algal communities may be N saturated in the absence of high NO_3^- loading because of high NH_4^+ loading. In these lakes, denitrification will be the primary NO_3^- sink and S_{NO_3} values will not be sensitive to changes in NO_3^- loading.

The effects of increasing acidification and NO_3^- deposition on R_{NO_3} will be a function of their effects on denitrification, nitrification and algal consumption rates. Denitrification appears unaffected by low pH (Rudd et al. 1986). On the other hand, Rudd et al. (1988) reported that nitrification ceased in two experimentally acidified lakes (Lakes 223 and 302S) at pH 5.4–5.7. Decreased NO_3^- formation probably contributed to lower NO_3^- export and, hence, to the high R_{NO_3} observed for these lakes. Increases in NO_3^- concentration have been observed in acidified lakes (Bowland, Lohi & Clearwater Lakes) in Ontario (Yan & Dillon 1984; Molot et al. 1990).

A similar analysis of lake retention of NH_4^+ shows that $1/R_{NH_4}$ was linearly related to $\bar{z}/\tau$ (Fig. 5b) with $r = 0.91$ and $S_{NH_4} \pm 95\%$ confidence limits $= 5.6 \pm 4.8\ m\,yr^{-1}$ for $1/R_{NH_4}$ versus $\bar{z}/\tau$ (Table 5). Correlation was best for $1/R_{TIN}$ versus $\bar{z}/\tau$ with $r = 0.91$ and $S_{TIN} \pm 95\%$ confidence limits $= 4.3 \pm 4.5\ m\,yr^{-1}$ (Fig. 5c).

Potential for spurious self-correlation exists in analysis of N retention in lakes (Table 6). Good correlation exists between retention and several variables which are component variables of retention such as areal N loading, flushing rate and $\bar{z}/\tau$ but low correlation exists between retention and variables such as mean depth or catchment area which are not component variables. Although NO_3^- retention and loading were highly negatively correlated, NO_3^- loading and $\bar{z}/\tau$ were also highly correlated ($r = 0.98$). Covariance among the three variables obscures whether retention is affected by loading or $\bar{z}/\tau$.

The importance of spurious self-correlation (Kenney 1982) was gauged by comparison with correlations from a randomly generated data set. The resulting correlation coefficient between randomly generated $1/R$ and $\bar{z}/\tau$ was 0.03, and the linear regression r^2 (coefficient of determination) was 0.001. Correlation between retention and loading was weak with $r = 0.14$ as was the correlation between loading and $\bar{z}/\tau$ ($r = -0.11$). The subset of data with $R > 0$ and lake outflow < (stream inflow + precipitation) yielded a correlation coefficient between $1/R$ and $\bar{z}/\tau$ of 0.10. Hence, we can conclude that spurious self-correlation was not significant in this relationship and that loading and/or $\bar{z}/\tau$ exerts a real and significant effect on N retention in lakes.

Table 6. Correlation coefficients between several variables for Ontario lakes listed in Table 5 (excluding L302).

	R_{TIN}	$\bar{z}/\tau$	R_{NO_3}	R_{NH_4}
Areal TIN loading	−0.90	+0.99	-	-
Areal NO_3^- loading	-	+0.98	−0.83	-
Areal NH_4^+ loading	-	+0.71	-	−0.74
τ	+0.85	-	+0.87	+0.74
$\bar{z}/\tau$	−0.91	-	−0.87	−0.89
A_d	−0.21	+0.33	−0.26	−0.10
A_o	+0.44	−0.42	+0.03	+0.46

Area loading $= \Sigma\,[N]_i q_i/A_o + \Sigma\,[N]_{ppt}\mathrm{PPT}$

Volumetric loading $= (\Sigma\,[N]_i q_i + \Sigma\,[N]_{ppt}\mathrm{PPT}\,A_o)/V$

Flushing rate, τ $= V/\Sigma\,q_0$

$\bar{z}/\tau$ $= \Sigma\,q_0/A_o$

$$R = 1 - \frac{\Sigma\,[N]_0 q_0}{\Sigma\,[N]_i q_i + \Sigma\,[N]_{ppt}\mathrm{PPT}\,A_o}$$

where R lake N retention
- $[N]_i$ N concentration in inflow streams
- $[N]_0$ N concentration in outflow streams
- $[N]_{ppt}$ N concentration in precipitation
- q_i volumetric discharge via stream inflow
- q_0 volumetric discharge via outflow
- PPT precipitation depth
- A_o lake area
- V lake volume
- $\bar{z}$ lake mean depth
- τ lake water replenishment time
- A_d catchment area

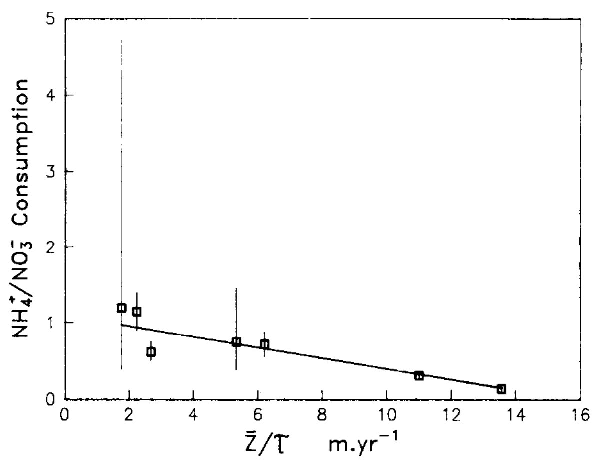

Fig. 6. Ratio of mean consumed NH_4^+ mass to mean consumed NO_3^- mass in lakes versus $\bar{z}/\tau$. Acidification occurs when the ratio is > 1. The line was fit with ordinary least squares regression using data from Table 6. Error bars indicate ranges.

Table 7. Ratio of NH_4^+ mass to NO_3^- mass consumption in several Ontario lakes. Net *N* consumption-based acidification occurs when the ratio of consumed NH_4^+ to consumed NO_3^- is > 1.

Lake	Year	NH_4^+/NO_3^-	Lake	Year	NH_4^+/NO_3^-
Harp	1976/77	0.94	Clearwater	1977/78	0.90
	1977/78	0.45		1978/79	1.40
	1978/79	1.46	Mean		1.15
	1979/80	0.90			
	1980/81	0.39			
	1981/82	0.56			
	1982/83	0.81			
	1983/84	0.46			
Mean (± SD)		0.74 ± 0.36			
			Lohi	1977/78	0.55
Plastic	1979/80	0.50		1078/79	0.88
	1980/81	0.75	Mean		0.72
	1981/82	0.59			
	1982/83	0.69			
	1983/84	0.58			
Mean (± SD)		0.62 ± 0.10			
Lake 239	1971	0.39			
	1972	1.79			
	1973	1.03	Batchawana	1981/82	0.34
	1974	1.06	South	1982/83	0.28
	1975	0.54	Mean		0.31
	1976	0.98			
	1977	0.71			
	1978	4.72			
	1979	1.14			
	1980	0.72			
	1981	0.60			
	1982	1.05			
	1983	0.86	Turkey	1981/82	0.09
Mean (± SD)	1971–83	1.20 ± 1.11		1982/83	0.19
Excluding	1978	0.91 ± 0.37	Mean		0.14

Effect of NH_4^+ and NO_3^- consumption on acidification

N-related acidification results not only from the presence of protons associated with unconsumed NO_3^-, but also from protons released upon NH_4^+ consumption. The relative effect of NH_4^+ and NO_3^- consumption on acidification of lakes is indicated by the ratio of their net consumption in lakes. This is true because loss of NH_4^+, whether it is via algal assimilation into biomass, or nitrification, results in the production of an almost equivalent amount of strong acid (in the latter case, after correcting the NO_3^- budget), and loss of NO_3^- by denitrification

Table 8. Ratio of NH_4^+ to NO_3^- consumption in several Ontario catchments. Net N consumption-based acidification occurs when the ratio is > 1. c/l is total catchment including lake.

Lake	Catchment									
		1976/77	1977/78	1978/79	1979/80	1980/81	1981/82	1982/83	1983/84	Mean
Harp	3	0.66	0.92	1.00	0.67	1.18	1.01	0.85	0.70	0.87
	3A	0.68	> 1	1.23	0.76	6.37	2.09	1.14	0.87	–
	4	0.61	0.71	0.98	0.68	0.82	0.63	0.74	0.64	0.73
	5	0.60	0.72	0.96	0.68	0.85	0.72	0.78	0.83	0.77
	6	0.68	1.06	1.10	0.76	0.97	0.82	0.95	0.80	0.89
	6A	0.64	0.72	0.92	0.64	0.78	0.63	0.70	0.62	0.71
	c/l	0.66	0.73	1.04	0.71	8.62	0.71	0.79	0.68	1.74
Plastic	1				0.64	0.77	0.63	0.71	0.63	0.68
	c/l				0.61	0.77	0.62	0.71	0.62	0.67
Clearwater	1		0.72	0.63						0.68
	2		0.73	0.64						0.69
	4		0.63	0.60						0.62
	c/l		0.73	0.69						0.71
Lohi	c/l		0.71	0.69						0.70
Batchawana South	c/l						0.91	0.83		0.87
Turkey	c/l						1.05	0.98		1.02
Kejimkujik	c/l				0.35					
		1971	1972	1973	1974	1975	1976	1977	1978	1979
Lake 239	NW	0.38	1.64	0.99	1.05	0.85	1.21	0.88	1.24	1.41
	c/l	0.38	1.66	1.01	1.09	0.86	1.28	0.86	1.43	1.37
		1980	1981	1982	1983	Mean				
	NW	0.85	2.24	1.19	0.82	1.13				
	c/l	0.86	1.34	1.18	0.82	1.09				
		1976	1977	1978	1979	1980	1981	1982	1983	Mean
Lake 223	c/l	1.22	0.85	1.32	1.32	0.85	1.33	1.11	0.81	1.10

or assimilation results in the production of an equivalent amount of base. This assumes that retained NH_4^+ is not adsorbed. If the ratio of consumed NH_4^+ mass to consumed NO_3^- mass is > 1, then acidification of the lake via consumption is occurring. As can be seen from the data in Table 7, net N consumption-based acidification occurred in 8 of 34 lake-years.

Mean net N consumption-based acidification was highly negatively correlated with $\bar{z}/\tau$ (r = 0.90, Fig. 6) and was more likely to occur in lakes with long residence times (i.e. when $\bar{z}/\tau < 1.5\,\mathrm{m\,yr^{-1}}$). Use of the regression equation in Fig. 5 is constrained by the assumption that NH_4^+ and NO_3^- loadings are not independent, although the large ranges for net N consumption-based acidification within a lake suggest otherwise.

Net N consumption-based acidification of soils occurred in the catchments of Turkey Lake, Harp 3A, 3 and 6, Lake 239 and Lake 223 (Table 8). The same phenomenon has been observed elsewhere. Extremely high levels of NH_4^+ deposition and high nitrification rates in the Netherlands have resulted in pronounced soil acidification (van Breemen et al. 1982).

Conclusions

The ability of forested stream catchments and lakes to retain NH_4^+ and NO_3^- loads was determined by mass balance calculations. However, differing system characteristics precluded direct comparison of retention between systems because of the effects of these characteristics on retention. Therefore, searching for evidence of 'NO_3^- saturation' by comparing systems in high and low deposition zones will be misleading unless relevant system characteristics are taken into account. Hence, analysis of synoptic survey data will be facilitated by grouping lakes and catchments according to similar characteristics.

If retention mechanisms are unaffected by changes in deposition, changes in deposition will be reflected by similar proportional changes in surface water concentrations assuming constant levels of N in internal storage pools. Therefore, increasing NO_3^- concentrations occurring simultaneously with higher deposition does not constitute evidence of saturation without reference to retention. For example, evidence of N saturation was found in southern Norway in two national surveys 9 years apart (Henriksen & Brakke 1988). TIN deposition increased 10% while surface water concentrations of NO_3^- increased approximately 200%. 'NO_3^- saturation' should, therefore, be defined as decreasing long-term, average NO_3^- retention in streams and lakes in response to long-term, average increases in NO_3^- (and NH_4^+) deposition.

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