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Assessing the Recovery of Lakes in Southeastern Canada from the Effects of Acidic Deposition

Reductions in North American sulfur dioxide (SO_2) emissions promoted expectations that aquatic ecosystems in southeastern Canada would soon recover from acidification. Only lakes located near smelters that have dramatically reduced emissions approach this expectation. Lakes in the Atlantic provinces, Quebec and Ontario affected only by long-range sources show a general decline in sulfate (SO_4^{2-}) concentrations, but with a relatively smaller compensating increase in pH or alkalinity. Several factors may contribute to the constrained (or most likely delayed) acidity response: declining base cation concentrations, drought-induced mobilization of SO_4^{2-}, damaged internal alkalinity generation mechanisms, and perhaps increasing nitrate or organic anion levels. Monitoring to detect biological recovery in southeastern Canada is extremely limited, but where it occurs, there is little evidence of recovery outside of the Sudbury/Killarney area. Both the occurrence of Atlantic salmon in Nova Scotia rivers and the breeding success of Common Loons in Ontario lakes are in fact declining although factors beyond acidification also play a role. Chemical and biological models predict that much greater SO_2 emission reductions than those presently required by legislation will be needed to promote widespread chemical and latterly, biological recovery. It may be unrealistic to expect that pre-industrial chemical and biological conditions can ever be reestablished in many lakes of southeastern Canada.

INTRODUCTION

Although a great deal is known about acidification of aquatic ecosystems in southeastern Canada (1, 2), much less is known about their chemical and especially biological recovery as acid inputs subside. Uncertainty extends to both the regional extent of recovery and the biogeochemical factors that influence or control it.

'Acid rain' gained prominence as an environmental issue in Canada during the 1970s when losses of fish species in the La Cloche Mountain (Killarney) lakes of Ontario were linked to acidification caused by atmospheric deposition of pollutants presumed to originate from nearby base metal smelters at Sudbury (3). Subsequent measurements of precipitation chemistry showed that acidic deposition did occur throughout southeastern Canada (4), and soon after, regional lake acidification was also confirmed (5, 6). Acidification was generally confined to the sensitive terrain (silicate bedrock overlain by thin to absent glacial deposits) found throughout the region—much of it called the 'Canadian Shield' (7) (Fig. 1).

The scientific consensus on the aquatic effects of acid rain that developed in North America during the 1980s prompted political action to reduce sulfur dioxide (SO_2) emissions (summarized in 8) which culminated in the 1991 Canada-US Air Quality Agreement (AQA, 9). Total North American emissions are now ~ 40% less than in 1980 (10). Over half of the eastern Canadian emission reductions have occurred at the base

metal smelters in Sudbury, Ontario and Rouyn-Noranda, Quebec. While both countries have monotonically reduced emissions for over two decades, the timing of the maximum rate of reduction differed between them being greater in Canada during the 1980s than the 1990s, and greatest in the USA during the mid-1990s.

Precipitation SO_4^{2-} concentrations in eastern Canada and the US have shown a general downward trend over the last two decades (11) leading to a decline in wet deposition in eastern North America. In Canada, southern and central Ontario and southwestern Quebec have undergone the largest reductions in regional SO_4^{2-} deposition. Most of southern Ontario and southwestern Quebec which received > 20 kg wet SO_4^{2-} ha^{-1} yr^{-1} (some areas were > 30 kg ha^{-1} yr^{-1}) in 1980–1984 received < 20 kg ha^{-1} yr^{-1} by 1995–1998 (12). Based on catchment mass balances (13), areas directly influenced by the Sudbury (and presumably Noranda) smelters have experienced even greater reductions in total acidifying deposition, much of it most likely as reductions in dry SO_2 deposition. A marked decrease in wet SO_4^{2-} deposition occurred across eastern North America in 1995 due to SO_2 emission reductions mandated in the USA by the 1990 Clean Air Act Amendments (requirements that were subsequently included in the AQA). The decrease is illustrated in Figure 2. Current deposition levels are still well above the critical load (usually ≤ 13 kg wet SO_4^{2-} ha^{-1} yr^{-1} and sometimes < 6 kg ha^{-1} yr^{-1}, see ref. 2) in sensitive regions of eastern Canada, prompting another domestic policy initiative to reduce SO_2 emissions even further (14).
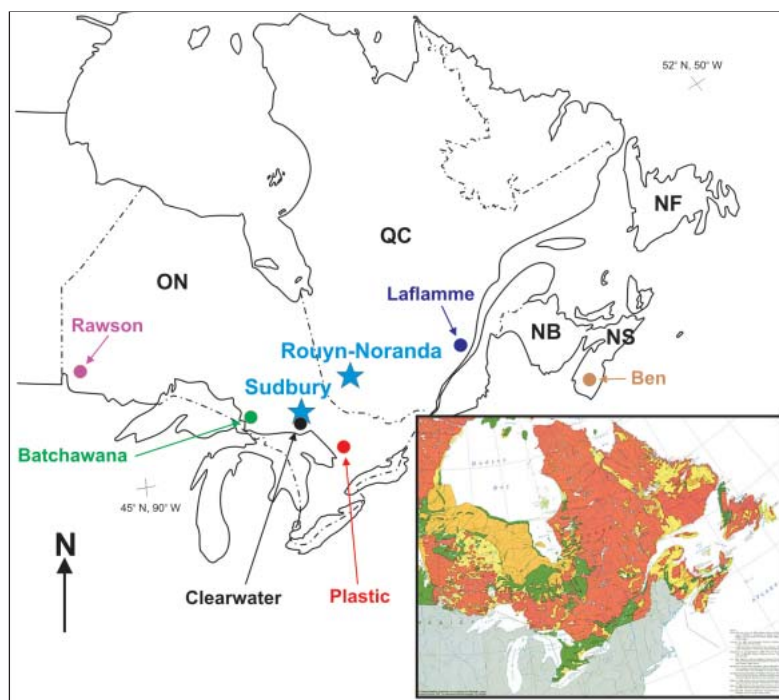

Figure 1. Map of southeastern Canada showing the location of provinces, smelters and the 6 lakes mentioned in the text (provincial abbreviations: Newfoundland = NF, Nova Scotia = NS, New Brunswick = NB, Quebec = QC, and Ontario = ON). Red coloration in the inset map denotes terrain that is sensitive to acidic deposition based on bedrock and soil characteristics (7).

Figure 2. Regional-scale non-sea-salt wet SO$_4^{2-}$ deposition and wet NO$_3^-$ deposition across eastern North America for the 1992–1994 and 1995–1997 periods. The decrease between the earlier and later periods was considerably greater for SO$_4^{2-}$ than NO$_3^-$, a reflection of the large reduction in SO$_2$ emissions in 1995 *versus* a minimal change in NO$_x$ emissions.

Nitrogen oxide (NO$_x$) emissions are also potentially acidifying. While the nitrate (NO$_3^-$) concentrations observed in some surface waters suggest that nitrogen saturation (and hence nitrogen-based acidification) is a developing issue in some ecosystems (15), SO$_4^{2-}$ deposition is still considered by far the predominant regional-scale agent for acidifying waters in southeastern Canada. Compared to SO$_2$, eastern North American NO$_x$ emissions changed little in the 1990s, resulting in minor changes to the wet NO$_3^-$ deposition patterns before and after 1995 (Fig. 2).

The declining deposition of acidifying SO$_4^{2-}$ in southeastern Canada prompted expectations of ecosystem recovery. This paper will address those expectations by summarizing changes in the chemistry and biology observed in and predicted for lakes across southeastern Canada, and by discussing factors that influence recovery. For the purpose of this paper, increasing pH or alkalinity trends in lakes will be taken as evidence of progressing chemical recovery.

Observations of lake chemistry changes were reported soon after SO$_2$ emissions and SO$_4^{2-}$ deposition declined (e.g. 16, 17). Selection of the time window for analysis was subsequently shown to have a profound effect on trend identification (18) underlining the fact that the time period discussed here (approximately two decades or less) is still relatively short for detecting/characterizing recovery in ecological terms. The literature therefore contains trend conclusions that commonly varied with time as dynamic lake ecosystems responded to changing acid inputs. Hence, this paper will give only the important generalities from older trend analyses and detail a compilation of trend results from the 1990s so as to establish a regional context for the analyses of the Killarney lake recovery presented elsewhere in this volume.

CHEMICAL RECOVERY

Trends near Smelter Sources

With particularly early and large (> 90%) emission reductions occurring at Sudbury, many papers have reported changes in lakes chemistry in the surrounding region. Such studies (e.g. 16, 19–21) produced a generally consistent picture of predominantly decreasing SO$_4^{2-}$, base cation, aluminum and base metal concentrations, and increasing pH and/or alkalinity (e.g. see Clearwater Lake in Figure 3). The rate (slope) of the SO$_4^{2-}$ changes was often steeper in the 1970s and 1990s than in the 1980s, apparently reflecting the SO$_2$ emission history at local smelters. The lake population being monitored and the time window being analyzed influenced the nature of the reported recovery response. For example, 36 of 38 relatively large lakes (14.5 to 1315 ha) moni-
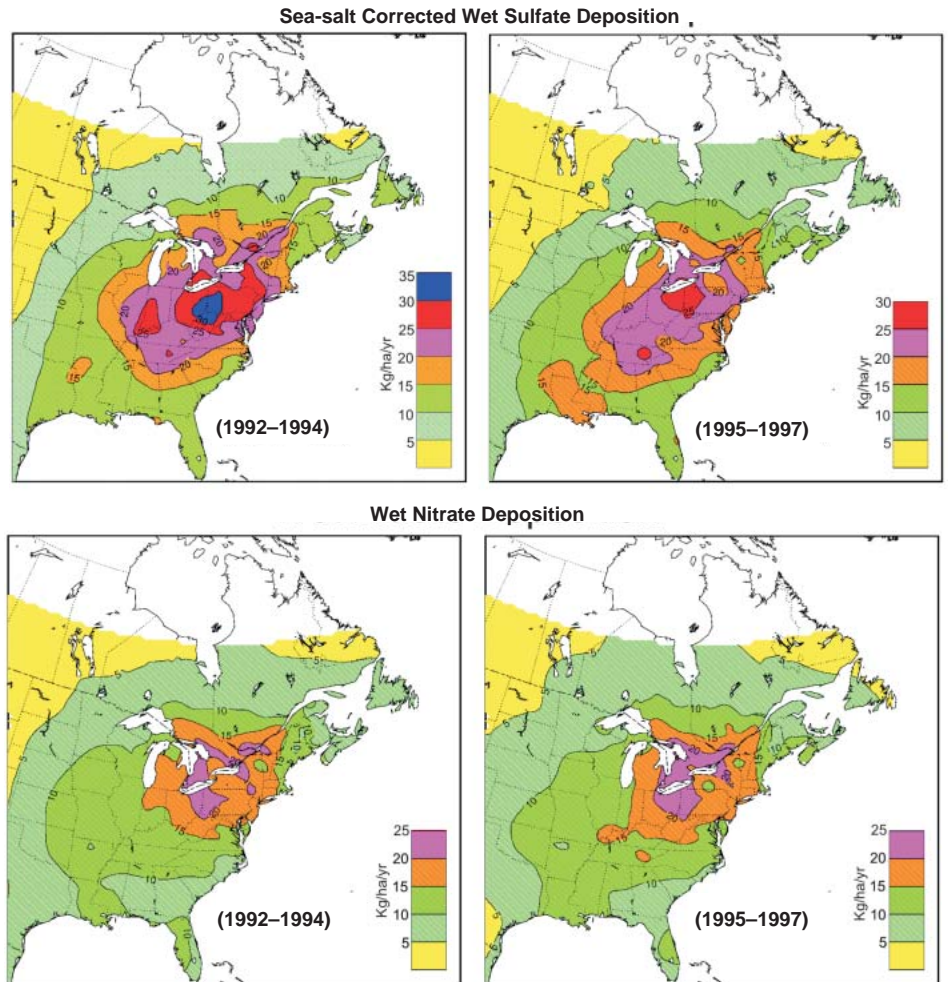
Figure 3. Annual average SO$_4^{2-}$ and pH for 6 lakes in southeastern Canada. Lake locations are shown in Figure 1.
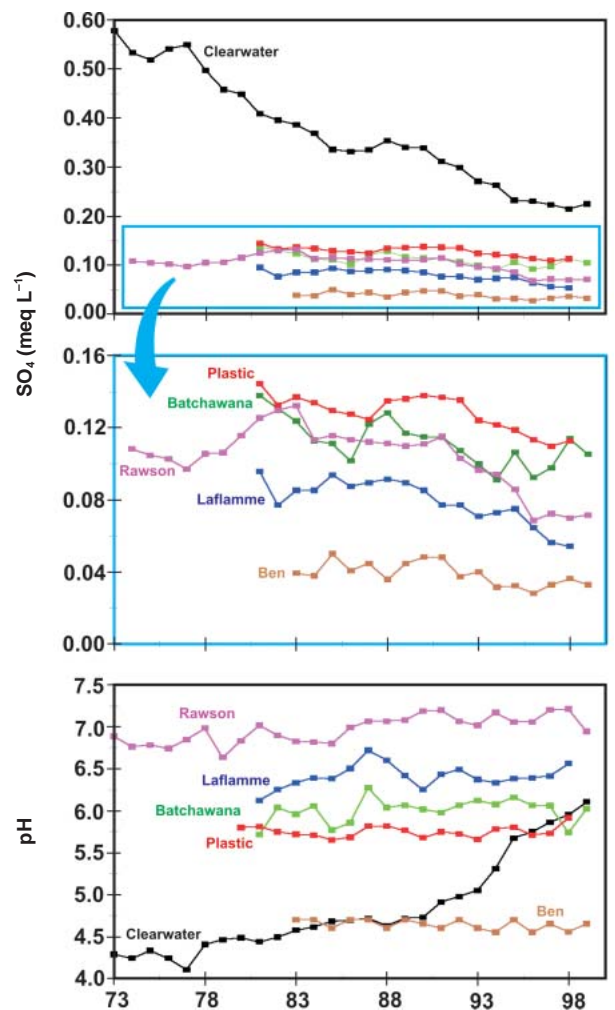
tored by the Ontario Ministry of the Environment (MOE) exhibited increasing pH between 1981 and 1989 (19), while only 26 of 161 smaller lakes (0.1 to 350 ha) sampled by the Canadian Wildlife Service (CWS) had increasing pH between 1983 and 1995 (2 were decreasing and the rest showed no change) (20). The potential increase in lake pH and/or alkalinity has been partially reduced by declining base cation concentrations (22). Specific departures from these recovery trends were usually readily explained, e.g. short-term, drought-induced trend reversals (19, 23; see later discussion). Despite the remarkable improvements of Sudbury area waters, several studies (e.g. 20, 21) still conclude that many lakes remain too acidic (pH < 6) to support sensitive aquatic biota.

The smelter located at Rouyn-Noranda is responsible for a large portion of Quebec's SO_2 emissions. An emission control program began in the early 1990s (much later than at Sudbury) with a 90% reduction goal (relative to 1980) by 2010. The current reduction level is ~ 70%. Water quality was measured in 64 lakes surrounding Rouyn-Noranda in 1982, 1991 and 1996 (24), and 48 lakes were re-sampled in 2001. The lakes are of 2 types: those located to the south and east of Rouyn-Noranda on the Laurentian Highlands are deep, clear, and sensitive to acidification, while those located on the Abitibi Lowlands near and north of the city are shallow lakes rich in dissolved organic carbon (DOC). Based on the 1982 results, the zone of direct smelter influence extends ~ 125 km from the smelter. Mean SO_4^{2-}, base cation and aluminum concentrations significantly declined in these lakes between 1982 and 1996, while alkalinity, NO_3^- and DOC-related variables increased. No significant pH increase was detected, however. Ten clearwater lakes within the zone of direct smelter influence were added to the 1991 and 1996 surveys. The mean pH of these lakes increased from 5.42 to 5.84 over the 5-yr period. The results suggest that chemical recovery is proceeding but incomplete for clearwater lakes and may be delayed for colored lakes. The instances of increasing NO_3^- are of particular interest because there has been no increase in nitrogen deposition or land-use changes to explain them, and they are spatially restricted to a zone within 50 km of the smelter. Increasing NO_3^- trends observed elsewhere in Quebec were more numerous in the 1980s than the 1990s (17, 25, 26).

Trends in Regions Principally Influenced by Long-range Sources

Atlantic Provinces
Analysis of monotonic SO_4^{2-} trends for the 1983–1997 period for 63 lakes in Newfoundland and Nova Scotia showed 34 cases of significantly (p ≤ 0.05) declining concentrations (27)—a shift from an earlier analysis of the 1983–1991 records that showed many cases of increasing SO_4^{2-} (17). 'No change' was by far the predominant response for pH, alkalinity, and base cations. These results agreed with those obtained using a more sophisticated statistical procedure on a combined Maine/Atlantic Canada dataset (25).

Lake surveys in New Brunswick were repeated irregularly 5 times between 1986 and 2001 (28). Evaluation of the chemical changes occurring between adjacent samplings generally corroborated observations made in the other Maritime provinces. Similar to observations from a more broad-based study (29), significant changes in water chemistry occurred only within acid-sensitive lakes in the survey (alkalinity < 40 µeq L^{-1}). Declining lake SO_4^{2-} was accompanied by decreasing pH and alkalinity between 1986 and 1993 due to sharp Ca^{2+} declines. Thereafter, Ca^{2+} levels stabilized or increased which led to increasing pH and alkalinity.

Quebec
Analysis of the 1983–1991 records for 29 lakes in southwestern Quebec showed that declining SO_4^{2-} was a predominant

chemical response (17). There were no cases of pH/alkalinity recovery, nearly equal numbers of increasing and decreasing base cation trends, and 11 increasing NO_3^- trends. A re-analysis with 8 additional lakes using the 1985–1993 sampling period confirmed the SO_4^{2-} response, but showed changes in other variables (26). Nine of the 37 lakes had significant increases in pH and/or alkalinity in this analysis. In addition, there were no cases of increasing base cations (17 were declining), fewer increasing NO_3^- trends (five), and 17 lakes exhibited decreasing DOC trends. These results corroborated the trends identified in a Quebec-Vermont dataset (25).

Ontario
All Ontario lakes from the 1983–1991 analysis of Canadian datasets (17) and the broader analysis of North American regions (25) covering the 1980s through to 1995 were located outside Sudbury's influence. These studies suggested that decreasing SO_4^{2-} and base cation trends became more prevalent and/or steeper in magnitude in Ontario from the 1980s to the 1990s, while trends for other variables were few or inconsistent between sites. Data from repeated surveys of 56 headwater lakes in Algoma (30) indicated that initial improvements in their acidity (median pH increased 0.5 units between 1979 and 1985) were lost by 1994 (2). The CWS monitored 216 and 235 small waterbodies in the Muskoka region (southeast of Sudbury) and the Algoma region (west of Sudbury) respectively (31). The CWS focus on small waterbodies (median size for the 2 datasets was 4.2 and 3.8 ha, respectively) yields sample populations that are more representative of the regions' actual lake population than other monitoring networks (2). For the CWS Muskoka and Algoma datasets, 47–56% and 51–57% of the lakes showed significant declining trends for SO_4^{2-} and base cations, respectively, between 1988 and 1996. Increasing pH and alkalinity trends occurred in only 12% and 21–23% of the lakes however.

Recent Trends (the 1990s)

For this paper we have compiled and/or determined monotonic trend information by province for all of southeastern Canada that approximately covers the 1990s (Table 1). All trends were identified using non-parametric test procedures selected according to the presence or absence of seasonality and/or auto-correlation in the datasets (31, 32). Two observations are ubiquitous, namely that there are virtually no increases in either SO_4^{2-} or base cations (just 1 and 3 such cases, respectively, out of a possible 798 cases). During the 1990s, declining lake SO_4^{2-} was a predominant trend (i.e. occurring in > 50% of monitored lakes) in all provincial datasets except Ontario. 'No trend' for SO_4^{2-} was still an important occurrence (41%) in Newfoundland lakes. Only 33% of monitored lakes in Ontario showed declining SO_4^{2-} mostly due to the lack of a monotonic response of small lakes and wetland water bodies in the CWS dataset. When the CWS data were excluded from the Ontario data, 80% of the lakes showed declining SO_4^{2-}.

'No trend' was the predominant response for base cations in Newfoundland and Nova Scotia during the 1990s (Table 1), presumably due to exhaustion of bases in the extremely thin soils that occur throughout the area. In contrast, declining base cation trends were very important in Quebec and Ontario. Once again, there was a great difference in the distribution of base cation trend direction in Ontario depending on whether the CWS data were included or excluded (inclusion resulted in similar levels of 'no' and decreasing trends while exclusion showed almost all decreasing base cation concentrations).

There is little evidence of chemical recovery in the Atlantic provinces. Only 10–14% and 0–5% of lakes in Newfoundland and Nova Scotia exhibited increasing pH and alkalinity, respectively. Most lakes showed 'no trend', and in fact, 3% and 9% of Nova Scotia lakes had decreasing pH and alkalinity; i.e. they

were still acidifying. A much higher proportion of lakes from southern Quebec were recovering; 37% and 49% had increasing pH and alkalinity, respectively. Even so, several lakes were still acidifying (12% and 19% had decreasing pH and alkalinity). While > 50% of Ontario lakes showed no recovery trend during the 1990s whether or not the CWS data were included in the evaluation dataset, there were still many cases of increasing pH and/or alkalinity. Small lakes in wetland landscapes had a lower incidence of recovery. When the CWS data were included in the Ontario set, 13% and 15% of the lakes exhibited increasing pH and alkalinity trends, respectively, while 39% and 41% had increasing trends when the CWS data were excluded. When acidifying trends (decreasing pH or alkalinity) were identified in Ontario, the proportion was higher in the dataset that excluded the CWS lakes.

Changes in organic anions (A^-, reflected in DOC concentrations) and NO_3^- may also influence recovery. 'No trend' in DOC concentrations was by far the dominant observation in Quebec and Ontario lakes, although there were some instances of both decreasing and increasing levels (Table 1; note that no DOC trend analysis was possible for the Atlantic provinces). As mineral acid deposition decreases, the influence of organic acids will increase due to their amphoteric nature, even without an increase in DOC (36). Nitrate was not included in Table 1 because in many datasets there are numerous occurrences of 'detection limit' data that make trend analysis problematic. Previous analyses did not find significant NO_3^- trends during the 1990s in eastern Canadian regions (25, 29). However, the CWS dataset of small Ontario lakes analyzed here showed 17% with increasing NO_3^- concentrations and 83% with no trend. Nitrate levels in these lakes are low (~ 1.2 μeq L^{-1} on average) meaning that even when they are increasing (mean positive trend was 0.28 μeq L^{-1} yr^{-1}), they are unlikely to greatly affect the acidity status in the near future.

Predictions of Chemical Recovery

Whether or not the degree of North American SO_2 emission reductions will permit regional chemical recovery has been evaluated using an integrated assessment model (2, 37). The model predicted that Canadian emission controls will reduce damage in Ontario and Quebec, but have little effect in Atlantic Canada. Implementation of further US SO_2 controls by 2010 will further reduce damage throughout all regions. Nevertheless, it was estimated that of 646 000 acid sensitive lakes in southeastern Canada (or 7 978 000 ha of sensitive lake area, 38), ~ 76 000 lakes (or ~ 970 000 ha) will remain chemically damaged unless additional SO_2 reductions are implemented beyond AQA requirements. Note that a 'damaged' lake was defined as having pH < 6 when deemed capable of a pH 6 or greater.

FACTORS INFLUENCING CHEMICAL RECOVERY

Trends in chemical recovery do not necessarily directly conform to trends in acidic deposition as shown above. There are many reasons.

In many Canadian lakes, declining base cations compensate for part or all of the declining SO_4^{2-} concentrations instead of increasing pH and/or alkalinity (2, 20, 22, 35), a phenomenon also observed in Europe and the USA (25, 29). In such cases, the lakes are not only failing to recover, but they are becoming more dilute. There are even a few cases where lakes with declining SO_4^{2-} continue to acidify at least in part because the rate of base cation decline exceeds the rate of SO_4^{2-} decline (35, 39). Depletion of the pool of easily exchanged base cations contained in drainage basin soils by acidic mobile anions seems to be the primary mechanism (40). However, other factors such as the glacial origin of Canadian Shield soils, soil thickness and profile maturity, occurrence of wetlands, declining base cation deposition (41) and other external stressors like climatic and land-use changes modify the base cation-SO_4^{2-} relationship. Replenishing the soil pool of exchangeable base cations via primary weathering will be a slow process, meaning that lake recovery may be slow (40), and in fact, it may be unrealistic to expect that pre-industrial chemical conditions can ever be re-established. Paleolimnological reconstruction of long-term Ca^{2+} patterns in 6 Sudbury-area lakes showed that concentrations have been declining since pre-industrial times (22).

The time required for lakes to re-equilibrate to lower SO_4^{2-} deposition differs from lake to lake. Lakes with long water replenishment times obviously take longer to reach a new steady-state than those with short replenishment times. However, calculation of SO_4^{2-} mass budgets show that even when accounting for the effect of water replenishment, terrestrial basin (stream) inputs and lake output generally exceed SO_4^{2-} deposition (42). Additional sources of SO_4^{2-} in the terrestrial basin are indicated, for example SO_4^{2-} desorbed from forest soils, SO_4^{2-} produced by oxidation and mobilization of reduced S stored in wetlands, stream margins, etc. and occasionally, SO_4^{2-} produced by oxidation of sulfide minerals present in the bedrock. These sources depend on spatially variable basin characteristics such as topography, bedrock geology, forest type, soil type, depth and chemistry, percentage of wetlands, etc. Major watershed disturbance may also play a role. The SO_4^{2-} increase that occurred in Rawson Lake at the Experimental Lakes Area (ELA) during the late 1970s and early 1980s (Fig. 3) followed 2 forest fires.

Table 1. Percent of monitored lakes in provinces of southeastern Canada (Fig. 1) having increasing, decreasing or no statistically significant[1] concentration trends during the 1990s.

Province, Period, Data source, No. lakes	Trend direction	pH (%)	Gran alkalinity (%)	SO_4^{2-} (%)	Base cations[2] (%)	DOC (%)
Newfoundland, 1989–1997 Clair et al. (27) n = 29	increase	10	0	0	0	NA[3]
	decrease	0	3	59	7	
	no trend	90	97	41	93	
Nova Scotia, 1989–1997 Clair et al. (27) n = 64	increase	14	5	0	2	NA[3]
	decrease	3	9	84	23	
	no trend	83	86	16	75	
Quebec, 1990–1997 Kemp (33) n = 43	increase	37	49	0	0	14
	decrease	12	19	98	70	10
	no trend	51	33	2	30	76
Ontario, 1990–1999[4] various sources[5] n = 662	increase	13	15	1	1	4
	decrease	1	1	33	47	5
	no trend	86	84	66	52	91
Ontario, 1990–1999, excluding CWS datasets, n = 54	increase	39	41	0	0	22
	decrease	7	2	80	98	4
	no trend	54	57	20	2	74

[1] Non-parametric trend test significance p ≤ 0.05 for all datasets except Quebec (p ≤ 0.10).
[2] Base cations comprised of sum of sea-salt corrected Ca^{2+}, Mg^{2+}, Na^+ and K^+ for Newfoundland and Nova Scotia, and the sum of Ca^{2+} and Mg^{2+} for the remaining provinces.
[3] DOC data from Newfoundland and Nova Scotia were not tested because of a mid-period change in analytical methodology known to give higher concentration values.
[4] 1990–1999 period common to all Ontario datasets (see footnote 5) except the CWS Algoma subset (1988–1999) and one ELA lake (1990–1997).
[5] Ontario data composed of: (a) Canadian Wildlife Service (CWS) unpublished datasets (n = 608 lakes) supplied by D. McNicol (cf. McNicol et al. 31), (b) Ontario Ministry of Environment unpublished data for Dorset lakes (n = 8) supplied by P. Dillon, (c) Sudbury unpublished long-term monitoring data (n = 38) supplied by W. Keller (cf. Keller et al. 22, 34), (d) Turkey Lakes Watershed unpubl. data (n = 2) supplied by D. Jeffries (cf. Jeffries et al. 35), and (e) Experimental Lakes Area (ELA) unpubl. reference lake data (n = 6) supplied by M. Turner.

The subsequent SO_4^{2-} decline reflects the deposition trend at ELA. Whether or not the movement towards new SO_4^{2-} input-output equilibria translates into discernible recovery trends also depends on the magnitude of the deposition reduction. Where it is large, e.g. near Sudbury and Rouyn-Noranda, both lake SO_4^{2-} and pH change quickly (e.g. Clearwater Lake in Figure 3) leaving little doubt that recovery is occurring as a direct response. Where the magnitude of deposition reduction is smaller (i.e. elsewhere in southeastern Canada), the other factors influencing the input-output equilibrium may obscure chemical trends and probably delay recovery for years if not decades.

Drought has particularly influenced SO_4^{2-} export from Ontario lakes (23, 35, 43). Watertable or lake-level draw-down allowed wetland soils and/or littoral zone sediments to dry thereby producing conditions where previously reduced S species could be oxidized. When the systems re-wetted, export of the resulting mobile SO_4^{2-} has produced both occasions of episodic acidification when H^+ was an important co-exported cation, and extended periods of elevated SO_4^{2-} (relative to pre-drought levels) when Ca^{2+} was typically a more important compensating cation. Increases in the annual average SO_4^{2-} concentration during the mid-1970s in Clearwater Lake and during the late 1980s and late 1990s in Batchawana, Clearwater, and Plastic Lakes (Fig. 3) were all attributed to the drought effect (8). Drought events delay lake recovery and work is underway to quantify the size of the reduced S pool and its accessibility in order to assess the long-term potential of this stressor interaction.

As SO_4^{2-} deposition declines, the relative importance of acidification by N deposition increases. Between 1982 and 1992, Lac Laflamme, Quebec, had declining alkalinity despite the fact that the SO_4^{2-} trend changed from increasing to decreasing around 1986 (Fig. 3). This alkalinity trend was attributed to the combined effect of declining base cations and increasing NO_3^- (39). In the Turkey Lakes Watershed in Ontario (cf. Batchawana Lake in Figure 3), increasing NO_3^- was observed in ground water and some surface water sites although its influence on acidity status was minor (35). At both Lac Laflamme and Batchawana Lake, increasing NO_3^- was probably more related to changing forest condition (and perhaps climatic variation) than atmospheric deposition. As noted above, no such explanation is appropriate for the NO_3^- increases observed in lakes near the Rouyn-Noranda smelter nor the numerous instances present in the CWS dataset. Whether or not N deposition in southeastern Canada is restraining lake recovery remains an open question.

Factors that affect internal alkalinity generating processes may profoundly influence lake acidification or recovery. The importance of internal alkalinity generation in lakes generally increases with the water replenishment time. Typically the most important processes underlying internal alkalinity generation include SO_4^{2-} reduction, denitrification, ion exchange at the water-sediment interface (44), and DOC degradation (P.J. Dillon, pers. comm.). Whole-lake acidification studies conducted at the ELA have indicated that internal alkalinity generation can be impaired such that the pH trajectory during recovery differs from the acidification trajectory (2). At a minimum there is a lag in the pH recovery relative to the SO_4^{2-} decline, and a significant likelihood that the chemical condition of the lakes is proceeding to a different state than the original (cf. movement to more dilute systems noted above). The reasons for the impairment of internal alkalinity generation in the ELA experiments remain speculative, perhaps being related to reduced availability of organic carbon to power the microbially driven SO_4^{2-} reduction. Depletion of the iron needed to form insoluble sulfide compounds has also been shown to reduce the internal alkalinity generation (45). Impairment of internal alkalinity generation also appears to depend partly on the severity of the acidification. For example, reducing the pH below 5 as was the case in one ELA experiment impaired the lake's internal alkalinity generation, while lowering the pH to only 5.1 in another experiment did not. The impairment of a lake's biogeochemical ability to internally buffer acids means that the efficiency of acidifying inputs is greater during the recovery period than it was during acidification.

BIOLOGICAL RECOVERY

With the exception of central Ontario, particularly the Sudbury/Killarney region, information on regional recovery in southeastern Canada is very limited. The reader should look elsewhere in this issue for discussion of the Sudbury/Killarney evidence. Monitoring specific biological groups in other regions offers some evidence (see below), but a regional biomonitoring network (46) that would have assisted assessment was terminated in 1997 before any biological trends were apparent. Simulation models have been used to predict biological recovery for scenarios of emission/deposition reduction. Detailed information on the processes and interactions that influence the rate and pathway of biological recovery has come from the whole-lake acidification/de-acidification studies conducted at ELA in northwestern Ontario. Such information is beyond the scope of our paper however. In this issue, information from ELA recovery experiments can be found for phytoplankton communities (47; see also 48) and epilithic algal assemblages (49), while information for fish has been published elsewhere (e.g. 50, 51).

Regional Monitoring

Nova Scotia is the most heavily impacted Canadian province in terms of the proportion of fish habitat that has been damaged by acid rain. There used to be 65 rivers originating from the Southern Upland with Atlantic salmon (*Salmo salar*). In 14 rivers that now have pH < 4.7, salmon have been extirpated. Twenty-four of the the rivers with pH from 4.7 to 5.0 are considered 'partially impacted', and 22 rivers with pH ≥ 5.1 are categorized as 'low impacted' (52). There is no evidence that the reduction in SO_4^{2-} deposition experienced thus far in Atlantic Canada is promoting recovery. Low marine survival is compounding the effects of freshwater acidification.

The Common Loon (*Gavia immer*), a conspicuous and popular fish-eating bird on boreal lakes, is symbolic of Canada's wilderness. As a top predator on lakes across eastern Canada during the summer months, the Common Loon has been used to indicate the extent to which acid-sensitive lakes and their food chains are recovering from acidification. Because two adult loons require up to 180 kilograms of fish to fledge one chick, breeding success can be lower on acid lakes where young may starve from lack of food (2). Loon breeding success has been monitored by CWS and volunteer surveyors across southeastern Canada (53, 54). In Ontario, temporal patterns for loon breeding on 292 lakes have been evaluated from 1987 to 1999. Reproduction was deemed 'successful' if a breeding pair was estimated to have fledged at least one young. The mean proportion of 'successful' pairs varied significantly over the 12-yr period, ranging from 0.26 to 0.77, and when the effects of lake area and pH were taken into account, there was a significant negative trend overall (Fig. 4). Although the overall importance of pH to loon breeding success was confirmed (55, 56), trends in success did not differ among pH classes. Other factors (perhaps mercury, climate change, ultraviolet radiation, and recreational development; 57–59) may have helped drive the downward trend.

Predictions of Biological Recovery

Assessment models have been used to predict whether aquatic populations in regions of southeastern Canada will eventually recover. Residual damage was evaluated relative to the predicted pre-acidification or 'potential' condition. Modelling taxonomic richness and occurrence across 7 groups of aquatic biota for a SO_4^{2-} deposition scenario reflecting full implementation of AQA

emission reductions suggested, no doubt conservatively, that ~ 25 000 lakes will still have a ≥ 20% deficit in potential richness (60). Ontario regions were predicted to recover to a greater extent than either south-central Quebec or southern Nova Scotia (54). Fish and mollusk were predicted to be the most affected groups, rotifers the least. Focusing on fish alone, predictions of residual damages of potential populations have ranged from ~ 133 000 populations (61) to ~ 162 000 populations (2, 54) across all southeastern Canada. Models that simulate the occurrence of loons and common mergansers (*Mergus merganser*) suggest that substantial improvements in habitat quality will only occur with SO_2 emission reductions beyond the AQA level (54, 56). Some assessments conclude that as much as an additional 75% in emission reductions will be needed to re-establish chemical conditions suitable for sensitive aquatic biota (14, 54).
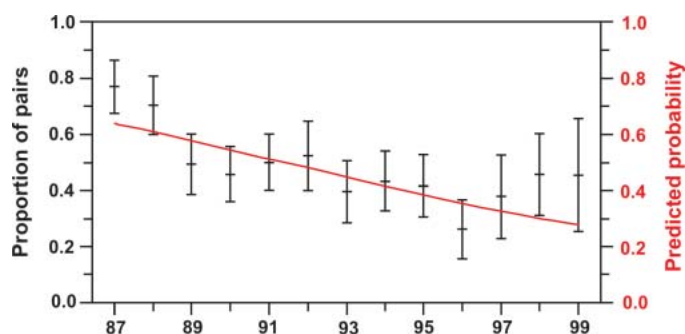
CONCLUSIONS

Lakes located near smelters that have reduced SO_2 emissions by ~ 90% show strongly declining SO_4^{2-}, distinct but incomplete trends in chemical recovery (increasing pH or alkalinity), and early stages of biological recovery. In the remainder of southeastern Canada, however, chemical recovery is progressing more slowly and is less well developed. The pace of biological recovery is slower than chemical recovery and less well understood.

Contrary to earlier expectations, declining SO_4^{2-} deposition does not necessarily mean immediate increases in lake pH or alkalinity. Regional data from the 1990s show that SO_4^{2-} is generally declining in lakes, but this is only now becoming commonplace in the Atlantic provinces and has been partially nullified in Ontario by drought-induced mobilization of SO_4^{2-} from landscape elements that are particularly important to small lakes and wetlands. While increasing lake pH and/or alkalinity is becoming increasingly prevalent (relative to earlier assessments), the predominant acidity response is still 'no trend', primarily due to base cation declines compensating for the SO_4^{2-} declines. The combination of declining SO_4^{2-} and base cations means that the lakes are probably becoming more dilute. For this and other reasons, many lakes are likely to recover to a chemical state that differs from their pre-industrial condition. Increasing NO_3^- and A^-, and damaged internal alkalinity generation mechanisms may play a role in the lack of recovery of some lakes.

Biological groups or indicators from regions of southeastern Canada other than Sudbury/Killarney show little change or declining condition, rather than recovery. This conclusion is based on extremely limited information, however, since there is no systematic monitoring program to detect biological recovery outside of south-central Ontario. The conclusion seems reasonable though, given the incomplete chemical recovery and expectations that biological recovery will lag behind the chemistry. Aquatic recovery will have a time line of at least several decades, not years.

Integrated assessment models predict that the reduced acidic deposition, resulting from implementation of AQA-required SO_2 emission reductions in eastern North America, will not be sufficient to achieve chemical or biological recovery of aquatic ecosystems in southeastern Canada. Additional emission reductions by as much as 75% are indicated.

Compared to the scientific effort that quantified aquatic acidification during the 1980s and early 1990s, current efforts to define aquatic recovery have been minimal. As a result, the limited information available describing the current status and trends of lake chemistry and particularly biology represents a continuing, major challenge for understanding regional recovery. There are many factors and processes influencing recovery of aquatic ecosystems that we do not yet fully understand, e.g. potential of climate-landscape interactions to delay recovery, processes underlying nitrogen-based acidification, interruption of internal alkalinity generation, and geographic barriers to biotic re-invasion of chemically suitable lakes. Nevertheless, we do understand that further emission reductions are necessary if acid sensitive lakes in southeastern Canada are to recover first chemically and then biologically.

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Figure 4. Mean (± 2 standard errors) proportion of 'successful' breeding of Common Loon pairs observed on 292 Ontario lakes, 1987 through 1999. The red line represents a significant downward slope of modeled success. The observed and modeled patterns differ slightly because modeled values take into account the variation in average lake area and pH resulting from annual differences in the set of lakes actually surveyed (e.g. low pH lakes predominated in the 1996 sample set).

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