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The Effects of Acidic Deposition on Aquatic Resources in Canada: An Analysis of Past, Present, and Future Effects

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ENVIRONMENTAL SCIENCES DIVISION
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ENVIRONMENTAL SCIENCES DIVISION

THE EFFECTS OF ACIDIC DEPOSITION ON AQUATIC RESOURCES IN CANADA: AN ANALYSIS OF PAST, PRESENT, AND FUTURE EFFECTS

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EXECUTIVE SUMMARY

The objective of this report is to assess the impacts of acidic deposition on aquatic resources in eastern Canada. This report addresses five questions.

1. What are the size, location, and current chemical condition of aquatic resources in eastern Canada that are thought to be vulnerable to acidic deposition?

Recent aquatic survey data for Canada provide the basis for determining the number of lakes potentially vulnerable to acidification from atmospheric deposition. In eastern Canada, east of the Manitoba-Ontario border and south of 52°N and excluding the Great Lakes, about half of the more than 700,000 lakes have acid-neutralizing capacity (ANC) less than 50 $\mu eq\ L^{-1}$. Between 12,000 and 14,000 lakes, or 1.7% to 2.0% of the total population of lakes, are estimated to have a pH less than 5.0. Although the surveys from which these estimates were derived were not statistically designed to make regional population estimates, the survey results demonstrate the large number of sensitive lakes in eastern Canada.

2. What are the extent and magnitude of chemical changes in surface water of eastern Canada that can be attributed to acidic deposition?

Evidence from regional surveys shows that acidic deposition is largely responsible for the acid-base chemistry of surface waters. Comparison of sulfate deposition and lake water sulfate concentrations in Canada reveals that nearly all of the sulfate in lakes can be ascribed to atmospheric deposition. Analysis of major ion concentrations indicates that ANC has been replaced by sulfate over

broad areas of eastern Canada, resulting in greatly decreased ratios of ANC:(Ca + Mg). The regional patterns of ion ratios correspond closely to patterns of atmospheric sulfur deposition, indicating that SO_4^{2-} replaced ANC in areas receiving high levels of acidic deposition (wet deposition greater than 20 kg SO_4^{2-} ha^{-1} $year^{-1}$).

3. What evidence exists to support the hypothesis that biological changes in surface waters in Canada have resulted from chemical changes associated with acidic deposition?

The few historical records of species richness and relative abundance of fish in streams and rivers in eastern Canada show a decline in the number of species and a reduction in the relative abundance of selected species with an increasing H^+ concentration for pH less than 6. The only documented historical record of declines in fish species richness in Canadian lakes is for an area downwind of a point source of sulfur dioxide and metal emissions from a copper smelter. Statistical analysis of these data using a variety of methods suggests that acidification has contributed to the loss of fish species from these lakes.

Results of recent synoptic surveys of lakes and streams in eastern Canada show a trend of lower richness of phytoplankton, zooplankton, benthic macroinvertebrates, and fish species with lower pH. These results are consistent with acidification being the cause of lower species richness and of lower relative species abundance. Laboratory bioassays and in situ bioassays tend to support the conclusion that acidification is the cause. Although there are no documented adverse biological effects of episodic pH depressions in Canadian lakes and streams, declines in pH and increases in aluminum during snowmelt events have been reported, and bioassays show that these episodic changes in water chemistry can stress aquatic organisms and increase mortality. Thus, there is reason for concern about potential adverse biological effects of episodic increases in acidity on aquatic populations and communities in surface waters in Canada.

4. How many systems in eastern Canada will become acidic at various levels of acidic deposition and within what length of time?

Simulation modeling experiments produce regional estimates of potential changes in surface water chemistry and damage to fisheries as a result of acidic deposition. Model results show that substantial decreases in the extent of acidification are possible with reduced deposition. For example, model simulations suggest that changing the atmospheric loading from 27 to 18 kg SO_4^{2-} ha^{-1} $year^{-1}$ would decrease the number of acidic lakes in eastern Canada (pH less than 5) by 41% to 51% (3,000 to 16,000 lakes). The loading value of 18 kg SO_4^{2-} ha^{-1} $year^{-1}$ is far below values currently measured in much of eastern Canada and would require additional control measures to attain.

Models of fisheries damage in Ontario suggest that, at steady state, 1980 levels of deposition will decrease species richness, probability of presence, and production of fisheries by 4.3%, 5.7-8.9%, and 2.3%, respectively. Emission controls in Canada would bring about recovery of 35% of the lost species richness, roughly one-third of the "lost" probability of presence, and 20% of the lost production.

5. What is the rate at which lake chemistry and biology improve when deposition is reduced?

If inputs of SO_4^{2-} to an aquatic system are reduced, the natural processes of ANC generation will increase the ANC and pH of the system with the decline in SO_4^{2-} concentrations. The observed recovery of lakes in Sudbury and of rivers in Nova Scotia has been in immediate response to decreases in sulfur deposition. The critical factor controlling the observed recovery of these once-acidic systems is a decrease in SO_4^{2-} deposition.

Mitigation studies in Ontario and Nova Scotia suggest that, in the absence of metal contamination, biological recovery does occur, although the rate and extent are still unknown.

1. INTRODUCTION
(Robert B. Cook and John L. Malanchuk)

1.1 OBJECTIVES

The objective of this report is to assess the current knowledge of surface water acidification and its effects on aquatic biota in eastern Canada. We used a critical appraisal of recent research to draw answers to the following questions:

- What are the size, location, and current chemical condition of aquatic resources in eastern Canada that are thought to be vulnerable to acidic deposition (Sect. 2)?
- What are the extent and magnitude of chemical changes in surface waters of eastern Canada that can be attributed to acidic deposition (Sect. 3)?
- What evidence exists to support the hypothesis that biological changes in surface waters in eastern Canada have resulted from chemical changes associated with acidic deposition (Sect. 4)?
- How many aquatic systems in eastern Canada will become acidic at various levels of acidic deposition and within what length of time (Sect. 5)?
- What is the rate at which lake chemistry and aquatic biota improve when deposition is reduced (Sect. 6)?

The aquatic resources evaluated in this assessment are in that area of Canada east of the Ontario-Manitoba border and south of 52°N, excluding the Great Lakes, and encompassing part or all of the provinces of Ontario, Quebec, Newfoundland, Labrador, New Brunswick, and Nova Scotia (Fig. 1.1). Included in this region are an estimated 700,000 lakes covering an area of 160,000 km^2 (Kelso et al. 1986a; Sect. 2).

This report was commissioned by the U.S. Environmental Protection Agency (EPA) for the purpose of: (1) serving as an update on the state of knowledge on the effects of atmospheric deposition on aquatic systems in eastern Canada since the U.S.-Canada Memorandum of Intent on Transboundary Air Pollution (MOI 1983) and (2) providing a companion to the EPA Staff Paper/Special Assessment on the effects of acidic deposition on aquatic chemistry and biology in the United States (EPA 1987). We addressed the same questions and used the same general outline as the EPA Staff Paper, so that in principal one could make comparisons between the two countries on the state of knowledge of the effects of acidic deposition on aquatic resources. EPA phrased the questions and formulated the outline based on the research framework developed in the Agency's Aquatic Effects Research Program (AERP). Because the Canadian counterpart to AERP, the Long-range Transport of Air Pollution Liaison Office, is not addressing the exact same questions, the scientific information available to answer the questions may, to some extent, be more limited than that in the United States. However, sufficient research findings are available on the effects of surface water acidification in Canada, so that we could provide scientifically meaningful answers to these five questions with attendant caveats and estimates of uncertainty.

1.2 SCOPE OF THE ASSESSMENT

In general, we evaluate material published since the MOI (1983). Much of the new information on which this assessment is based comes from two sources:

- Proceedings of the International Symposium on Acidic Precipitation held in Muskoka, Ontario, September 15-20, 1985 (published in Water, Air, and Soil Pollution, Volumes 30 and 31, 1986) and

- Assessment of the State of Knowledge on the Long-Range Transport of Air Pollutants and Acid Deposition [Federal/Provincial Research and Monitoring Coordinating Committee (FPRMCC) (November 1986)]. FPRMCC (1986) also draws heavily on results presented in the Muskoka Proceedings.

This assessment is not limited to peer-reviewed, published information. The best and most recent publicly available information is used. Where the quality of the available information was unknown or poor, that information was either not used or was supplemented with appropriate warnings as to limitations of the data.

This assessment was not prepared in response to FPRMCC (1986). Rather this assessment was prepared as a companion to the EPA Staff Paper as described above. Two members of the FPRMCC (P. J. Dillon and K. H. Mills) critically reviewed this assessment prior to publication.

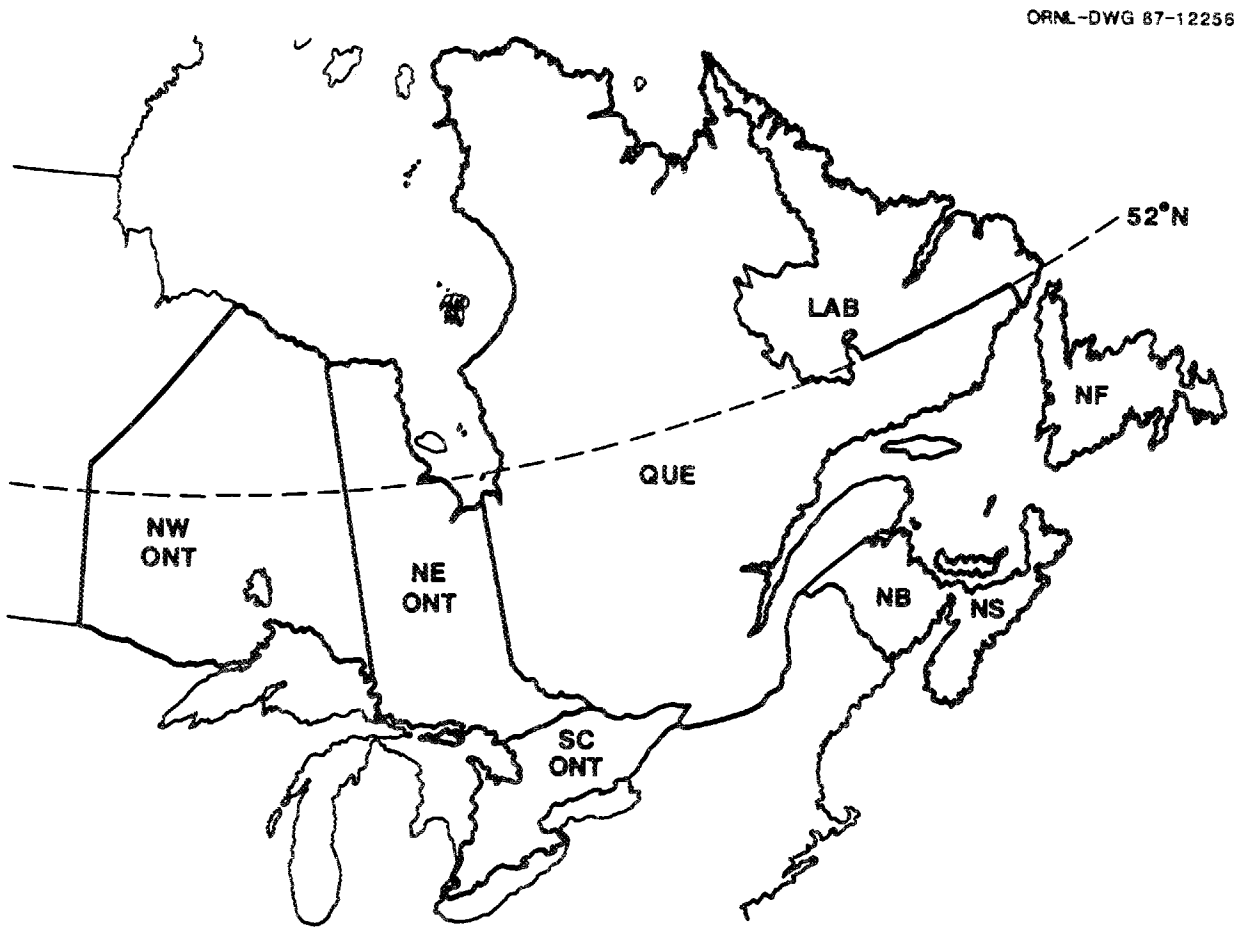

Fig. 1.1. Map of Canada, showing provinces and regions east of the Manitoba border. Source: Jeffries et al. (1986).

2. PRESENT STATUS OF SURFACE WATER CHEMISTRY
(Michael L. Jones)

In this section, estimates are presented on the size, location, and current chemical condition of aquatic resources in eastern Canada that are thought to be vulnerable to acidic deposition. The basis for these estimates is existing lake chemistry data, particularly those from extensive regional surveys of lakes in sensitive regions of eastern Canada. The focus is on lake water chemistry largely because in Canada existing data on surface water chemistry and biota are primarily for lakes and also because the aquatic resources at risk to acidic deposition are found predominantly in lakes, not running water. The processes contributing to the present chemical status of lakes in Canada are discussed later in Sect. 3.

The section begins with a definition of the area of interest. Then, the various sources of data on current status of lake water chemistry are identified, and the limitations of these data are discussed. Third, estimates of the overall size of the surface water resources are presented, without regard to their chemical status, after which the current chemistry of surveyed lakes is examined to determine the likely causes of the observed lake water chemistry. The resulting data are then combined into an overall extrapolated estimate of the number of low-ANC (acid-neutralizing capacity) lakes.

The survey data presented in this section focus exclusively on the water chemistry currently observed in lakes; these data are not sufficient to examine temporal variation in chemistry. However, the evidence for short-term, episodic changes in surface water chemistry is also considered. This evidence is limited and tends to be site specific; thus, it does not lend itself at the present time to regional extrapolation. Nevertheless, it illustrates the potential for the occurrence of significant, short-term changes in lake water chemistry at particular locations.

2.1 AREA OF INTEREST

This assessment considers eastern Canada, east of the Manitoba-Ontario boundary and south of 52°N (Fig. 1.1). Within this overall area, there are some regions in which soils and bedrock have a moderate-to-high potential for reducing the acidity of atmospheric deposition (Shilts et al. 1981), such as southwestern Ontario and most of Quebec south of the St. Lawrence River. These areas were generally not included in the surveys discussed below. On the other hand, the surveys do include some lakes in sensitive areas north of 52°N, most notably in Labrador. The survey data, therefore, do not exactly correspond to the area of interest but are instead better thought of as generally reflecting the chemistry of lakes in sensitive areas of eastern Canada. Because the lakes surveyed were not chosen to be statistically representative, extrapolation to the region of concern will contain uncertainties, as discussed below.

2.2 SOURCES OF DATA

2.2.1 Number of Lakes and Lake Area

The first step toward identifying aquatic resources at risk in Canada is to estimate or measure the size of the overall resource--in this case, the number or area of lakes in sensitive areas of eastern Canada south of 52°N. The only published estimate of the size of the low-ANC lakes in eastern Canada was made by Kelso et al. (1986a). Kelso et al. used the Canadian almanac (Bracken 1981) as their source for area of lakes and combined this with estimates of lake size distributions (Fig. 1 in Kelso et al. 1986a) obtained from analyses of topographic maps (see Minns 1984) to estimate the number of lakes. Since Bracken (1981) only provides estimates of freshwater area for entire provinces, Kelso et al. (1986a) had to assume that the area of surface waters south of 52°N is distributed in direct proportion to the area of land masses in each province to arrive at these estimates.

2.2.2 Water Chemistry

Over the past decade, several water chemistry surveys have been conducted in eastern Canada, most of which have focused either on individual provinces or on regions within provinces. The most notable exception is the National Inventory Survey (NIS) conducted by the Department of Fisheries and Oceans from 1979 through 1982, which included lakes from all provinces east of Manitoba, except Prince Edward Island. Detailed descriptions and analyses of these data are provided by Kelso et al. (1986a). For the most part, the results of other lake chemistry surveys have not been published. Recently, however, Jeffries (1986) and Jeffries et al. (1986) have compiled many of these data, including data on the 814 lakes in the NIS, into a single Canadian data base of more than 7000 lakes. Discussions of the current chemical status of lakes in eastern Canada will thus focus on these two data bases (Table 2.1).

2.3 LIMITATIONS OF DATA BASES

Kelso et al.'s (1986a) estimates of resource size for all of eastern Canada have to be considered highly uncertain. The Canadian almanac (Bracken 1981) is unlikely to be a source of precise estimates of the area of surface waters; the method of calculation is not documented. In addition, the lake size distributions are based on relatively small samples, especially in Quebec. Other unpublished studies have used different methods of estimating resource size (Jones et al. 1984: see Table 5.3 in this document; R. Helie, Environment Canada Land Directorate, personal communication to M. L. Jones, 1987) and have arrived at significantly different estimates, at least for Quebec.

Analytical methods for the NIS data base are described in Kelso et al. (1986a). All laboratories making analyses for NIS also participated in round-robin comparisons with other laboratories in the United States and Canada. The data were subjected to simple quality assurance checks, such as charge balance and computed-vs-measured conductivity comparisons, prior to statistical analysis. Clearly

Table 2.1. Number of lakes sampled by region for the National Inventory Survey (NIS) and the overall Canadian data bases and estimated total number of lakes

Region	Number of lakes sampled		Estimated total number of lakes south of 52°N
	NIS[a]	Overall Canadian[b] data base	
Nova Scotia	76	275-413	6,585
New Brunswick	69	52-81	3,596
Newfoundland	109	176-268	138,711
Quebec	251	1268-1452	441,752
Ontario	179	-	126,278
South-central Ontario		750-1578	
Northeastern Ontario		793-1805	
Northwestern Ontario		216-1078	
Total	684	3,530-6,675	716,922

[a]From Kelso et al. (1986a).
[b]From Jeffries (1986). Sample size for each province/subregion varies depending on chemical parameter of interest.

anomalous observations (e.g., errors due to incorrect data entry) were corrected whenever possible; those not correctable were excluded from subsequent analyses. Because lake SO_4^{2-} was measured using a colorimetric method (methylthymol blue), systematic positive biases may be present in these data for lakes colored by dissolved organic matter (Kerekes et al. 1984). Because such errors did not appear in the charge balance analyses, which included estimates for organic anions using the method of Oliver et al. (1983), SO_4^{2-} data from colored lakes were not excluded from the analyses presented in Kelso et al. (1986a).

Jeffries et al. (1986) describe the methods used to analyze the data included in their data base. In contrast to Kelso et al. (1986a), they exclude all colorimetric SO_4^{2-} measurements for lakes whose color exceeded 30 hazen units (or 6 mg L^{-1} dissolved organic carbon). They also describe the quality assurance protocols used to screen the data bases during and subsequent to chemical analysis.

Data used to create the data bases discussed here were not derived from statistically designed surveys (i.e., those surveys designed to provide a basis for extrapolation to the total population of lakes in an area). Therefore, the results of any attempt to use these data for extrapolation must be interpreted with caution. In Nova Scotia, for instance, the surveyed lakes were apparently concentrated in areas of particularly high sensitivity, so that population-level extrapolations for this area will probably provide overestimates of the resources at risk unless the appropriate corrections are made. The lack of an objective basis for such corrections necessitated the use of uncorrected extrapolations in the estimates of the number of lakes and their chemical status (Sects. 2.3 and 2.4); the development of improved estimates, using the appropriate corrections, should be a high priority in future research.

For the NIS data base, the percentage of lakes sampled is very low, especially for Ontario and Quebec (Table 2.1). The addition of provincial data improves the situation for these two provinces (Jeffries 1986) (Table 2.1), although the sampling rate remains low, and the coverage is far from spatially uniform. In Quebec, most of the

surveyed lakes are relatively far south, while in Ontario, sampling is concentrated in the northeastern and south-central regions.

To evaluate the representativeness of the survey data, it is also useful to consider the purpose for which they were collected. The NIS was designed to concentrate on headwater lakes. In contrast, the Ontario provincial data base tends to be biased toward important sport fishing lakes, which are typically larger, higher-order lakes. Comparison of the NIS data with those of Jeffries et al. (1986) (the latter being dominated by provincial data) for Ontario should indicate the magnitude of the biases in these data sets.

2.4 RESOURCE SIZE

Kelso et al. (1986a) estimated that there are more than 700,000 lakes in the area of interest for this assessment (Table 2.2). Over half (56%) of the estimated 161,890 km^2 of lake surface area in eastern Canada is in Quebec. Ontario has the second largest area of lakes, but insular Newfoundland is estimated to have a greater number of lakes, because of the much smaller average lake size in this province (i.e., many more small lakes).

2.5 CHEMICAL CHARACTERISTICS OF SURVEYED LAKES

The NIS and the overall Canadian data bases show broadly similar patterns of lake chemistry across regions of eastern Canada (Table 2.3). Base cation (Ca + Mg) concentrations consistently increase from east to west. The relatively low median base cation concentrations in all regions indicate that sensitive lakes in eastern Canada are typically dilute, low-conductivity systems. The strong correlation between ANC and (Ca + Mg) that exists in these lakes (see Fig. 8 in Kelso et al. 1986a) is also indicative of dilute, low-conductivity waters, as observed for many surface waters having ANC and (Ca + Mg) less than the world average concentration (1000 $\mu eq\ L^{-1}$ and 1000 $\mu eq\ L^{-1}$, respectively) (Henriksen 1982a).

Table 2.2. Freshwater lakes in eastern Canada

Province	Total freshwater area (km^2)[a]	Percentage south of 52°N[b]	Freshwater lakes south of 52°N[c]	
			Area (km^2)	No.
Ontario (excluding Great Lakes)	86,600	65.4	56,600	126,000
Quebec	184,000	49.1	90,300	442,000
Newfoundland/Labrador	22,700	48.2	10,900	139,000
New Brunswick	1,340	100.0	1,340	3,600
Nova Scotia	2,650	100.0	2,650	6,600
Total	297,290	54.5	161,790	717,200

[a]Bracken (1981).
[b]U.S.-Canada Memorandum of Intent on Transboundary Air Pollution (1983) (MOI 1983).
[c]The number of lakes is determined from the area of freshwater south of 52°N (column 1 times column 2) and an estimate of lake-size distribution obtained from topographic maps (Minns 1984).

Source: Kelso et al. (1986a).

Table 2.3 Median lake chemical characteristics by province and, for Ontario, by region, for the National Inventory Survey (NIS) and overall Canadian data bases

Region	pH		ANC ($\mu eq\ L^{-1}$)		Ca + Mg ($\mu eq\ L^{-1}$)		SO_4^{2-} ($\mu eq\ L^{-1}$)	
	NIS[a]	Overall[b]	NIS[a]	Overall[c]	NIS[a]	Overall[c]	NIS[a]	Overall[c]
Nova Scotia	5.23	5.21	2	10	42	53	64	50
New Brunswick	6.62	6.38	52	38	106	90	80	77
Newfoundland	6.09	6.18	38	28	57	79	36	31
Quebec	6.50	6.33	46	52	109	163	61	93
SC Ontario	6.00	6.31	41	63	194	199	171	158
NE Ontario	6.40	6.92	83	165	42	441	138	148
NW Ontario	6.60	7.10	142	254	241	221	96	75

[a]Data from the NIS (Kelso et al. 1986a) with medians calculated by M. L. Jones.
[b]From Jeffries et al. (1986), Table 1.
[c]From Jeffries (1986), Table 1.

In Nova Scotia, the NIS and overall data bases yield similar patterns of lake water chemistry. For both data bases, median pH, ANC, and (Ca + Mg) values are lower in Nova Scotia than those in any other region. Only Newfoundland lakes have lower median sulfate values. Although there has been some suggestion that the existing Nova Scotia lake survey data may be biased toward low-ANC lakes (C.K. Minns, Canada Department of Fisheries and Oceans, personal communication to M. L. Jones, 1987), the data presented in Table 2.3 indicate that lakes sampled in Nova Scotia are among the most sensitive in eastern Canada.

Newfoundland lakes also have relatively low ANC, with median values <40 $\mu eq\ L^{-1}$ for the two data bases. Sulfate concentrations are lower than those found in Nova Scotia (50 vs 31 $\mu eq\ L^{-1}$), and pH and ANC values are higher (6.18 vs 5.21 and 28 vs 10 $\mu eq\ L^{-1}$).

The surveyed lakes in New Brunswick and Quebec have remarkably similar median values. The median ANC and (Ca + Mg) for lakes in these two provinces are higher than those for lakes in Newfoundland or Nova Scotia. Sulfate medians are also somewhat higher in New Brunswick and Quebec than in Newfoundland and Nova Scotia. The Quebec survey data tend to be concentrated in the southwestern region of the province, north of the St. Lawrence River. The limited sampling of more remote areas in northern Quebec (e.g., the north shore of the Gulf of St. Lawrence) suggests that the lakes in these areas are more similar in chemistry to Newfoundland lakes than to southwestern Quebec lakes.

The lakes in the three Ontario regions have both the highest median (Ca + Mg) and the highest median sulfate concentrations of any of the provinces in eastern Canada (Table 2.3). Median sulfate concentrations in south-central and northeastern Ontario are the highest in eastern Canada (171 and 138 $\mu eq\ L^{-1}$, respectively). Although patterns in (Ca + Mg) for the three regions in Ontario are not evident in these data, there is a striking inverse relationship between median sulfate and pH or ANC. Sulfate concentrations are highest in south-central Ontario and lowest in northwestern Ontario; ANC and pH exhibit the opposite patterns (Table 2.3). As discussed in Sect. 3.4, these observations are consistent with the hypothesis that the acid-base chemistry of lakes is largely the result of atmospheric sulfate deposition.

For all regions except Ontario, the two data bases show similar patterns of median lake chemistry. The only significant exception to this is the (Ca + Mg) values for Quebec, which are significantly higher in the overall data base (109 vs 163 $\mu eq\ L^{-1}$). In Ontario, there are a number of significant differences between the two data bases. Median pHs and ANCs are consistently lower in the NIS. Sulfate and (Ca + Mg) values in the two data bases are similar, except for (Ca + Mg) in northeastern Ontario. Therefore, for the lakewater chemistry in Ontario, the NIS data appear to be biased toward more sensitive lakes than is the overall Canadian data base.

In Ontario, the NIS sampled small headwater lakes exclusively. In contrast, the overall data base for Ontario is dominated by provincial data that are biased toward larger lakes containing sport fishes. In Sect. 5, predictions of future impacts of acidic deposition on regional lake chemistry use the Ontario provincial data as the basis for regionalization. From the standpoint of biases in the data, these predictions are therefore expected to underestimate the resources affected by changes in atmospheric deposition.

2.6 EXTRAPOLATION TO OVERALL RESOURCES

Ideally, estimates of the number of lakes with low ANC would be based on a survey that had been statistically designed to represent the population of lakes of interest. Unfortunately, such a survey does not exist for Canadian surface waters. Nevertheless, to make an assessment, the best possible estimate of the present chemical status of the entire population of aquatic resources must be determined. Although extrapolations based on existing survey data must be considered questionable, as mentioned in Sect. 2.3, there are no compelling reasons to believe that these data are strongly biased such that they would lead to gross over- or underestimates of the resources at risk, at least on a national scale. For example, the estimates for Nova Scotia are now thought to be overestimates due to a bias in sampling towards more sensitive areas. However, this uncertainty for Nova Scotia has a relatively small influence on the national figures.

Kelso et al. (1986a) used the results of the NIS analysis to estimate the chemical status of aquatic resources in eastern Canada. They estimated more than 350,000 lakes in the area of interest to have ANCs less than 50 $\mu eq\ L^{-1}$, whereas more than 14,000 were estimated to be acidic, according to the Henriksen (1980) nomogram (i.e., pH <4.7). For comparison, Jones et al. (1984) calculated that 12,500 lakes had a pH <5, using a larger data set that is a subset of the Jeffries et al. (1986) data base (see Table 5.3). The estimate determined by Jones et al. (1984) is quite similar to the value of 14,000 determined by Kelso et al. (1986a). Differences between the two estimates are principally due to differences in the estimated regional resources (higher for Quebec, lower for Maritimes in Kelso et al. 1986a). Finally, Minns (1986) estimated the number of lakes in Ontario having a pH <5 and ANC <0 $\mu eq\ L^{-1}$ to be 1418 and 3208, respectively. Again, the estimates do not depart substantially from those reported by Kelso et al. (1986a) and Jones et al. (1984).

There are two key reasons for not discounting the magnitude of the estimates because of biases in the underlying sample from which they were made. First, even if the values are overestimates, the acidic resources in Canada (pH <5 or ANC <0 $\mu eq\ L^{-1}$) are far greater than are those in the United States. For example, 238 of the 7096 lakes have a pH <5.0, and 325 lakes in the northeastern U.S. have an ANC <0 $\mu eq\ L^{-1}$ (Linthust et al. 1986). Second, using a pH of 5.0 as a criterion for acidification effects on biota is probably a lower limit. Biological effects begin when pH drops to 6.0 and continue until loss of sport fishes at pH 5.0.

2.7 EPISODES

Since the MOI was published, a great deal of the attention of Canadian researchers has shifted from consideration of the effects of long-range transport of atmospheric pollutants on the average condition of surface waters to the potential for short-term, episodic changes in water chemistry, especially during the spring snowmelt. These episodes may result in chemical conditions far more toxic to aquatic biota than those conditions experienced on the average, and the timing and

location of episodes may overlap with particularly sensitive life history stages of some biota. Nevertheless, it should be pointed out that although numerous examples of episodic changes in surface water chemistry have now been documented (see Marmorek et al. 1984 and 1987 for extensive reviews), there is very little direct evidence of the toxicity of episodes on biota (Sect. 4.4). This lack of evidence may be a consequence of the practical difficulty of observing such effects, however, rather than an indication of their absence in natural systems.

Primarily for practical reasons, research in this area has concentrated on site-specific studies, particularly in calibrated watersheds in Ontario and Quebec (Turkey Lakes, Dorset area, Lac Laflamme). There are noteworthy exceptions, however. Wilson and Barrie (1981) used data on climatic conditions and sulfur deposition to develop a very simple regional model of snowmelt episodes for eastern Canada. Their model assumes that snowmelt flows over frozen soil directly into surface water without any watershed neutralization. Thus, the Wilson and Barrie model must be considered a worst-case analysis. Nevertheless, the model effectively illustrates which areas of eastern Canada are most likely to experience severe snowmelt episodes, strictly from the standpoint of geographical patterns of winter/spring climate and sulfur deposition. Briefly, their results indicate that sulfur deposition is more important for acidic episodes than is climate, although they do demonstrate some influence of climate in areas where deposition rates are similar. Their analysis does not include the Maritimes, however. In maritime regions where the climate is more moderate, storm events are more likely to lead to significant episodes than are snowmelt events, although the limited data available suggest that storm episodes are generally less severe than snowmelt episodes (Marmorek et al. 1987).

Kelso et al. (1986b) monitored the chemistry of 30 small headwater lakes in north-central Ontario during the spring snowmelt period of 1981. All 30 lakes experienced declines in ANC during the melt period, although the pH and ANC changes were not due to changes in SO_4^{2-}, which apparently decreased or remained constant during this period.

NO_3^- was not measured in the Kelso et al. (1986b) study. For the high-ionic-strength lakes in this study, decreases in pH and ANC during snowmelt were due to base cation dilution.

Numerous other studies have documented snowmelt- and storm-event-related episodes at individual sites. For example, Craig et al. (1986) observed a pH depression of 0.6 pH units (5.2 to 4.6) and an ANC decline of 25 $\mu eq\ L^{-1}$ (25 to 0) while SO_4^{2-} and NO_3^- increased in near-surface groundwater (soil water) during the snowmelt period at the Turkey Lakes Watershed in north-central Ontario. Stream and lake acidic episodes during snowmelt have also been documented in this area (Jeffries and Semkin 1983), although changes in pH and ANC were better correlated with changes in flow than with changes in SO_4^{2-} and NO_3^-. Declines in ANC and pH associated with increases in SO_4^{2-} and NO_3^- during snowmelt have been described at other Canadian sites as well: Muskoka-Haliburton, Ontario (Jeffries et al. 1979; LaZerte and Dillon 1984; Scheider et al. 1984) and Sudbury, Ontario (Gunn and Keller 1984). Scheider et al. (1979) also described a pronounced depression in stream water pH during stormflow at Harp Lake, Ontario. Declines in pH of up to 1 pH unit without changes in ANC can also occur under ice during winter due to the buildup of CO_2 (Schindler and Nighswander 1970; Kratz et al. 1987).

2.8 SUMMARY

The survey data are presented for eastern Canada, a region that corresponds to the area east of Manitoba and south of approximately 52°N. The aquatic resources in the area are estimated to be 716,922 lakes that have an area of 161,890 km^2. There is considerable uncertainty associated with this estimate.

The current chemical status of lakes was determined from two data bases containing data for some 7000 lakes. Neither of the two data bases was designed to provide a statistical basis for extrapolation to the total population of lakes in an area. Although different chemical analysis techniques were used within the data bases, the data have been screened for some quality assurance checks (e.g., charge balance).

Despite these problems, both data bases show broadly similar patterns of lake chemistry across regions of eastern Canada. Base cations, sulfate, and ANC values generally increase from east to west. Median lake sulfate concentrations are highest in south-central and northeastern Ontario, and ANC and pH are lowest in Nova Scotia, Newfoundland, and south-central Ontario.

Kelso et al. (1986a) estimated that more than 350,000 lakes in the area of interest have an ANC <50 $\mu eq\ L^{-1}$, and that more than 14,000 lakes have pH <4.7. Another study estimated that the number of lakes with pH <5.0 was 12,500 (Jones et al. 1984). The estimates are comparable in size; their uncertainty is unknown, however.

These estimates of water chemistry are for average lake conditions. Short-term episodes of low pH and ANC during spring snowmelt are evidently quite common, although little direct evidence exists of the impacts of these episodes on biota.

3. QUANTIFYING CHEMICAL CHANGE AS A RESULT OF ACIDIC DEPOSITION

(Michael L. Jones, Robert B. Cook, John P. Smol, and Robert S. Turner)

3.1 CONCEPTUAL FRAMEWORK

The objective of this section is to assess the extent and magnitude of chemical changes in surface waters that can be attributed to acidic deposition. The principal problem associated with quantifying the cause-effect and dose-response linkages between acidic deposition and changes in surface water chemistry is that long-term historical data are lacking. No complete record exists of changes in atmospheric deposition, of emissions of acidic deposition precursors, or of changes in surface water chemistry over the past 100 years. Therefore, we can only infer relationships between acidic deposition and surface water chemistry. For example, changes in the composition of the remains of algae and trace metals in the sediment of lakes can be used to infer changes in lake water chemistry and atmospheric deposition. Results of studies of lake sediment are discussed in Sect. 3.2.

A scientific consensus about the atmospheric, watershed, and aquatic processes that mediate surface water chemistry has emerged in Europe and North America over the past 10 years. Figure 3.1 identifies the primary processes thought to control surface water chemistry. They have been reported in numerous technical publications, and their interrelationships are summarized in publications such as NRC (1986), Turner et al. (1986), Cosby et al. (1985a), Driscoll and Newton (1985), Mason and Seip (1985), EPA (1985), Johnson et al. (1985), Altshuller and Linthurst (1984), Goldstein and Gherini (1984), NAS (1984), Galloway et al. (1983), Marmorek et al. (1987), and Jones et al. (1987). This conceptual framework of atmospheric-soil-water processes is the basis for process-level studies in watersheds and for models that mathematically link acidic deposition inputs to surface water chemistry changes. Section 3.3 discusses the process study results to date, including the limited existing short-term atmospheric deposition and lake chemistry measurements. Section 3.4 discusses the magnitude

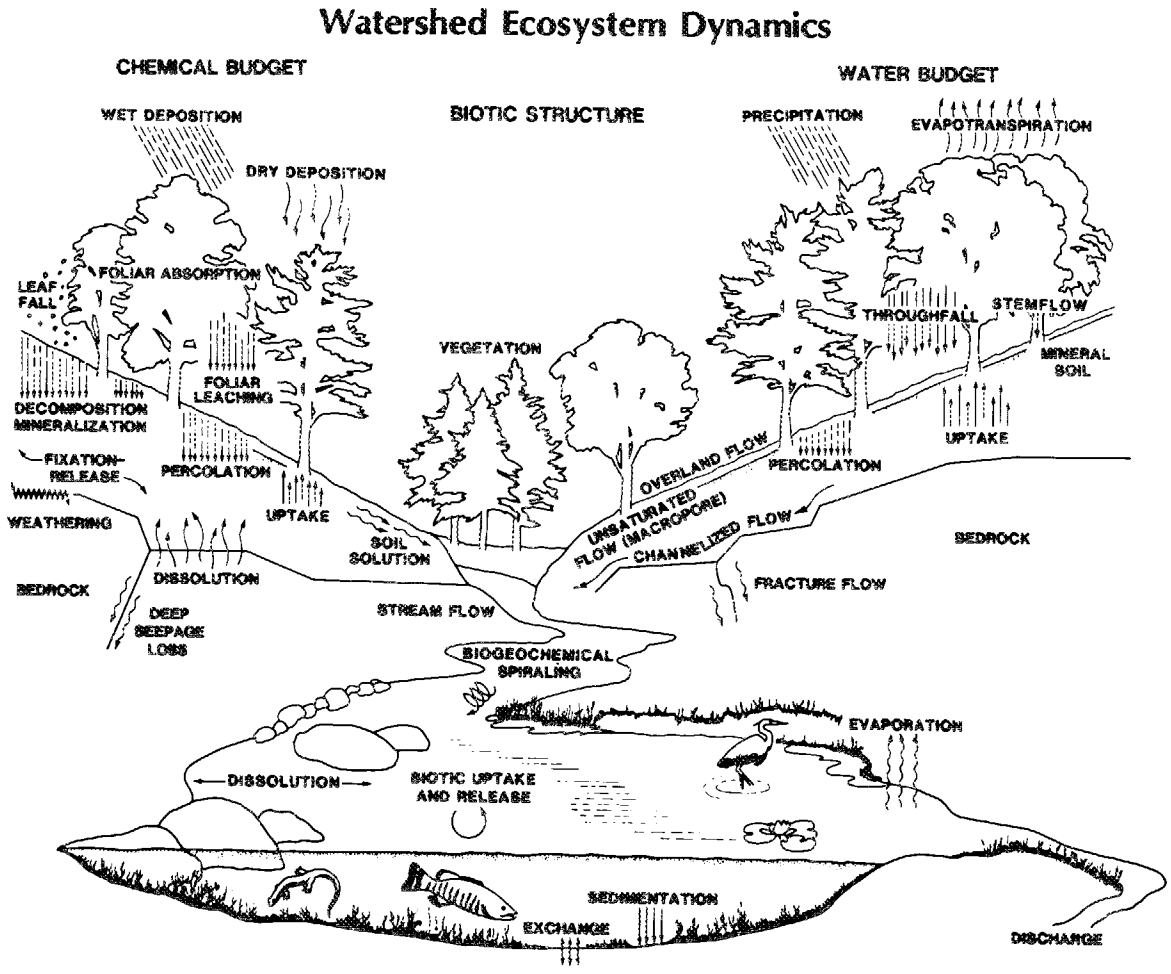

Fig. 3.1. Schematic diagram of a watershed, showing key hydrologic and biogeochemical processes that affect acidic deposition.

and extent of lake chemistry change, determined from empirical modeling using the existing extensive data on acidic deposition and lake water chemistry.

3.2 EVIDENCE OF LONG-TERM CHANGE AS A RESULT OF ACIDIC DEPOSITION

The evidence available to document changes in surface water chemistry is derived from two sources. Those sources are paleolimnological data, which provide a historical record of parameters that change along with water chemistry, and chemical data collected from lakes or streams over time.

3.2.1 Paleolimnological Evidence

Lacustrine sediments contain information on a lake's past: its biota, water chemistry, watershed characteristics, and the material deposited directly from the atmosphere. The information is in the form of organic and inorganic substances that entered or were formed within a lake and were deposited in its sediments. For the information contained in the sediments to be interpretable, the signal associated with the various substances cannot have been modified subsequent to deposition. Furthermore, to obtain a historical record of this information, the times when particular intervals of sediments were deposited need to be determined using radionuclide dating (^{137}Cs, ^{210}Pb), varves, or changes in sediment characteristics that correspond to well-dated local events (e.g., as evidenced by pollen or charcoal).

Sediment substances that provide the most evidence for long-term change as a result of acidic deposition are diatom and chrysophyte assemblages and trace metal content. A summary of findings for lakes in North America is presented in NRC (1986).

3.2.1.1 Diatoms and Chrysophytes

Diatoms (class Bacillariophyceae) are a large group of single-celled algae that are abundant in most lakes and that have siliceous cell walls. Chrysophytes (family Mallomonadaceae) are freshwater phytoplankton that have siliceous scales, spines, and bristles that can also be used for paleoecological reconstructions.

The relative abundance of specific taxa of diatoms and chrysophytes can be related to the lake water pH because most species are characteristic of restricted pH regimes. For diatoms, a calibration data set for a given region can be established using pH-preference categories or using multiple linear-regression of lake water pH with diatom species abundance in recent surface sediments (e.g., Smol et al. 1986). Because diatoms are relatively well-preserved in sediments, an analysis of their distribution and abundance in dated sediment layers may be used to reconstruct a quantitative estimate of historical trends. For chrysophytes, pH transfer functions have just been developed (Charles and Smol, in press). However, even without these quantitative equations, changes in depth profiles of chrysophyte assemblages can be interpreted in light of the known present-day preferences of chrysophyte taxa for certain lakewater pH ranges.

Reconstructions of pH histories of a number of lakes are now available (NRC 1986). These reconstructions show that acidification has occurred over time scales ranging from the past 5,000 years, attributable to natural processes for some lakes, to the past 50 years, attributable to long-range transport of acidic deposition (NRC 1986). For other remote regions, some lakes have apparently been acidic (pH <5) as a result of natural processes for up to 5,000 years (Whitehead et al. 1986; Kreis et al., in press).

To assess recent acidification trends in eastern Canada, data sets were selected that met the following criteria: (1) ANC $\leq$200 μeq L^{-1}, (2) minimal watershed disturbance, (3) sufficient quality diatom or chrysophyte data, and (4) sufficient quality sediment data.

Delorme et al. (1984) reconstructed pH trends using diatom assemblages for Kejimkujik Lake in central Nova Scotia, an acidic lake (pH 4.8) with high concentrations of dissolved organic matter. The authors concluded that acidic conditions have persisted in the lake for the last 1000 years, although assemblage changes in the upper few millimeters of sediments may possibly be attributable to anthropogenic acidification (H. C. Duthie, University of Waterloo, Waterloo, Ontario, personal communication to J. P. Smol, 1987).

Batchawana Lake, near Sault Ste. Marie, Ontario, has a present pH of 6.1 and ANC of 31 $\mu eq\ L^{-1}$ and apparently has undergone little pH change in the past 200 years (Delorme et al. 1986). The recent histories of three lakes in Algonquin Park (Ontario) that have pH values between 6.0 and 6.8 indicate that none had any post-1900 change in pH (Smol and Dickman 1981).

Hudon et al. (1986) examined the recent histories of two lakes in northeastern Quebec (Key Lake, pH 5.2; Lake C-22, pH 5.8). Although they were unable to infer a statistical change in pH because of a poor correlation between diatom taxa and pH, they were able to show a decrease in the number of indifferent taxa (distributed equally above and below pH 7) and an increase in the number of taxa that have an optimum distribution at pH values below 5.5.

Elner and Ray (1987) constructed diatom-inferred pH chronologies for three lakes in Nova Scotia and four lakes in New Brunswick. In Nova Scotia, one of the three low-ANC lakes showed a decline of pH from 6.1 to 5.3 over the period 1939 to 1979. The other lakes showed either no change or change within the uncertainty of the diatom technique. In New Brunswick, two of the four lakes studied revealed a decline of 0.5 pH units over the past 70 years.

Scruton et al. (1987) calculated diatom-inferred pH stratigraphies for seven lakes in insular Newfoundland. Three of the lakes have recent declines of 0.2 to 0.3 pH units that were attributed to increases in atmospheric deposition and to natural processes, either forest fires or production of organic acids. Two of the lakes showed no trend in diatom-inferred pH and two of the lakes exhibited an

increase in diatom-inferred pH. The magnitude of the pH changes observed are consistent with the relatively low rates of atmospheric deposition in Newfoundland and indicate that other factors in addition to atmospheric deposition are affecting the acid-base chemistry of the study lakes in Newfoundland.

Acidification has been inferred for lakes located near smelters in Wawa and Sudbury (Ontario). Lakes B and Cs have become more acidic in the past 30 to 50 years, apparently as a result of local sources of sulfur emission from Wawa that increased during the 1940s and 1950s (Dickman and Fortescue 1984; Dixit 1983; Dickman et al. 1984). In Hannah Lake (near Sudbury), acidification from pH 6.0 to pH 4.6 in 1975 apparently commenced with the start of smelting activities at Sudbury in the 1880s (Dixit 1986). In Clearwater Lake, (near Sudbury) the pH declined from 6.0 in about 1930 to pH 4.2 in 1970 (Dixit 1986). Because of the relatively slow sedimentation rate in Clearwater Lake, the rapid increases in pH that occurred with the decrease in emissions in the Sudbury area (0.4 units in 7 years; Sec. 6.2.1) are not resolvable (Dixit 1986). Due to the dominance of local sources, the pH trends for these four lakes cannot be used to help interpret changes in the long-range transport of strong-acids or their precursors.

Published paleolimnological data for eastern Canada, then, are insufficient to examine acidification of lakes attributable to the long-range transport of acidic deposition. Of the five lakes distant from local point-source emissions for which pH reconstructions have been made, none indicated acidification. Four of the lakes, however, are relatively insensitive to acidic deposition because they have pH and ANC values high enough to provide a pH buffer intensity against the levels of acidity associated with acidic deposition during the past 30 to 100 years. The fifth lake, Kejimkujik, has been acidic for the past 1000 years, apparently from organic acids generated in the watersheds.

In addition to the published literature, several research projects currently nearing completion are examining lakes in Quebec and Ontario that are remote from point sources, yet have a pH low enough to have

potentially resulted from acidic deposition. Dixit et al. (1987) used diatoms and chrysophytes in their paleolimnological study of 12 Quebec lakes located in a 150 km-wide strip north of the St. Lawrence River between the Ottawa and Saguenay rivers. In the more acid lakes, recent declines in lakewater pH have been inferred.

Another study currently nearing completion has used chrysophycean microfossils to infer pH changes from a variety of Ontario lakes (K. Nicholls, Ontario Ministry of the Environment, and J. P. Smol, unpublished data). In each of the presently acidic lakes studied, chrysophytes indicate that lakewater pH has declined in recent years. These sites include two lakes in Algonquin Park (Maggie and Pincher), two lakes in the Parry Sound region (Lady and Raven), and two unnamed lakes in Pukaskwa National Park (west of Wawa). The quantification of these pH declines is currently being assessed.

3.2.1.2 Geochemical Evidence

The chemical composition of lake sediments is the net result of all chemical inputs and outputs over time. Changes in atmospheric deposition can modify leaching rates of certain chemical species from the terrestrial environment, thereby affecting the input of material to the lake. Additionally, many constituents deposited from the atmosphere may be incorporated directly into accumulating sediments.

Because the acidity of precipitation in eastern North America is due in part to sulfuric acid, several studies have attempted to use the variation in sulfur content of the sediments as a surrogate indicator of the atmospheric deposition of sulfate (Mitchell et al. 1981, 1985). Sulfur may reach the sediment through a variety of pathways, including (1) sedimentation of allochthonous and autochthonous organic matter that contains sulfur; (2) sorption of sulfate on sedimenting detritus; and (3) reduction of sulfate in pore water or the water column, with subsequent precipitation of sulfides (Holdren et al. 1984). Because sulfur-containing phases can be chemically altered after deposition, the sulfur content of a particular interval of sediment may have no

direct relationship to the atmospheric deposition of sulfur. Comparison of the atmospheric deposition history inferred from sulfur emission estimates with the sulfur concentration profiles in lake sediments reveals no apparent cause and effect relationship (NRC 1986).

The sediment content of certain heavy metals may indicate atmospheric deposition from air masses polluted by human activity (NRC 1986). However, the sediment content cannot be directly related to the acidity of deposition.

The relationship between local point sources of pollution and enrichment of these various substances in sediments of adjacent lakes is well established. The classic example of a point source is the smelter at Sudbury, Ontario, that emits several pollutants associated with the processing of nickel and copper ore. Nriagu (1983), Nriagu and Wong (1983), and Nriagu et al. (1982) found that in dated cores from lakes proximate to the smelter the variations in concentrations for As, Se, Ni, Cu, Zn, and Pb paralleled the history of ore production at Sudbury. The source of these metals could be unambiguously identified as the Sudbury smelter.

Sediment chemistry from Lakes Tantare and Laflamme (Quebec, Canada) reveals that Pb and Zn concentrations increased in the period from about 1900 to 1980 (Ouellet and Jones 1983). Evans et al. (1983), Dillon and Evans (1982), and Evans and Dillon (1982) examined the trace metal content in sediments from 10 lakes in the Muskoka-Haliburton area. The chronology of the increasing concentrations found by Evans and Dillon (1982) is similar to that reported for lakes in New England and New York (NRC 1986).

The limited evidence from sediment chemistry profiles from Canadian lakes that are remote from point sources suggests a history of changing atmospheric chemistry over the past 100 years. This trend agrees with sediment data for the northeastern United States, which implicates changes in regional atmospheric pollution as the likely cause of changes in the trace metal content of sediments.

3.2.2 Historical Surface Water Chemistry

Comparisons of chemical data from surface waters that were sampled on two or more occasions many years apart may provide the best evidence for long-term changes in surface water chemistry. Such comparisons, however, have been hampered by the poor documentation of older records, the unreliability of older records, or the unreliability of older chemical methods. NRC (1986) has critically evaluated historical chemistry data for lakes and streams in the United States. The most recent summary of chemical changes in Canada's surface waters is the MOI (1983), which is the basis for this section of the report.

Watt et al. (1983) compared the major ion chemistry for several Nova Scotia rivers sampled in 1954 and 1955 and again in 1980 and 1981. SO_4^{2-} concentrations increased during this period (Watt et al. 1983). Even though organic matter interferes with the colorimetric SO_4^{2-} measurement, the change in measured SO_4^{2-} should be directly attributable to changes in SO_4^{2-}, if dissolved organic carbon (DOC) remains constant during this time period (MOI 1983). pH declined by 0.2 to 0.7 units between 1954-55 and 1980-81. Although pH was measured with a glass electrode in the earlier period, the uncertainty associated with these earlier measurements was probably $\pm$ 0.3 units. Thus, the pH measurement uncertainty obscures the magnitude of the pH trend.

Data from three rivers in Newfoundland (Isle aux Morts, Garnish, and Rocky) over the period 1960 to 1980 or 1981 indicate minimum discharge-weighted pH in 1972 or 1973 (Thompson et al. 1980). Wiltshire and Machell (1981) reported pH and ANC changes for 10 remote lakes in Nova Scotia and New Brunswick sampled in 1950 and 1979 and for which the most reliable historical information exists. These data indicate that calculated ANC declined by between 5.5 and 55 $\mu eq\ L^{-1}$ and that there has been a decline in pH for each of the lakes, although all but one of the 10 lakes had a pH >5.5 in 1979. The conclusions of these two studies rely on historical pH measurements that have a relatively large uncertainty.

For Clear Lake, in Central Ontario, an area of high sulfate deposition, Dillon et al. (1979) compared ANC data collected in 1977 with ANC data reported by Schindler and Nighswander (1970) and found a decline of 18 to 31 $\mu eq\ L^{-1}$. In northwestern Ontario, 11-year data sets for several lakes showed no decline in pH and calculated ANC due to precipitation of pH 4.9 to 5.0 (Schindler and Ruszczynski 1983).

Clair and Whitfield (1983) examined trends in pH, SO_4^{2-}, and Ca^{2+} over time (1960s through 1970s) in rivers in the Maritimes using time-series regression techniques. Trends in pH in rivers thought to be sensitive to acidic deposition were either decreasing (four rivers) or showed no change (three rivers). One of the rivers that showed no change (Mersey) had a mean pH of 4.9 during the study period and is not likely to be greatly affected. Another of the rivers showing no change (Lepreau) was studied for a short period, while the third (Piper's Hole) had the highest ANC of this sensitive group and would therefore be more resistant to pH change than the others. Calcium declined in three of the rivers and remained stationary in the other four, and sulfate declined in three of the seven. Clair and Whitfield (1983) conclude that although pH declined in some of the rivers, the pH decreases could not be linked to increases in sulfate.

Thus, the existing historical data are limited for the period of interest--the past 10 to 50 years. Historical data for SO_4^{2-} and NO_3^- are generally not available to document that the changes have been caused by acidic deposition. Changes in pH and ANC are consistent with acidic deposition-caused changes in surface water chemistry, although because of relatively high uncertainty in historical pH measurement techniques, the magnitude of the pH trend, if any, is not well established. Furthermore, in regions where paleoecological techniques have been used in conjunction with analysis of historical data (e.g., southern Ontario), similar conclusions have been reached--that acidification of lakes and streams occurred in geologically sensitive areas of Canada that receive acidic precipitation. However, the historical data are insufficient to address such issues as the extent and magnitude of aquatic resources that have acidified as a result of acidic deposition.

3.3 EVIDENCE FOR CAUSE-EFFECT RELATIONSHIPS BETWEEN ACIDIC DEPOSITION AND SURFACE WATER CHEMISTRY

3.3.1 Introduction

In Canada the consensus is widespread that long-range transport and the subsequent deposition of acidic substances have been responsible for significant changes in the surface water chemistry of northeastern North American lakes and rivers (FPRMCC 1986). Nevertheless, some controversy remains, especially over the extent of the present and anticipated future impacts of atmospheric deposition on surface waters. Proponents of alternative hypotheses (e.g., Rosenquist 1978; Krug and Frink 1983) suggest that there are important natural sources of acidity in watersheds and that these sources can be greatly influenced by land management activities within the watershed of interest. These authors suggest that further evidence is required to distinguish which of these hypotheses offers a better explanation of the current status of lakes and rivers in eastern Canada, with respect to acidification.

In Sect. 2, we presented data from Canadian surveys that provide some indication of the current status of Canadian lake chemistry. There is little doubt that the actual number of lakes, both those that are already acidic and those that could potentially become acidic, is substantial. The question to be addressed here is whether the observed current status of lakes in eastern Canada can be attributed in part to the effects of acidic deposition, particularly that resulting from the long-range transport of atmospheric pollutants.

To discuss this question we will draw upon three general sources:

1. detailed, intensive studies of catchment and in-lake processes that shed light on the potential for atmospheric-vs- catchment-derived processes to contribute to surface water acidification;

2. direct evidence for changes in lake chemistry as a result of changes in deposition, obtained from studies of lakes in the Sudbury area of Ontario; and

3. evidence from regional-scale analyses of survey data.

3.3.2 Process Studies

Processes occurring within the soils, vegetation, and surface waters of catchments can profoundly influence the chemistry of the surface waters. Not only are these processes thought to play a central role in mediating the effect of acidic deposition on water chemistry (as described in Sect. 3.1), but they can also be significant sources of acidity themselves. Several authors have shown that organic anions can contribute significantly to the acidity of surface waters (Kerekes 1977; Gorham et al. 1986; Underwood et al. 1986). Organic anions are almost exclusively derived from nonatmospheric sources. Thus, the acid-base chemistry of surface waters is governed by natural sources of acidity, along with other sources of acidity (e.g., atmospheric deposition) and sources of ANC. The issue is the relative importance of watershed-derived sources of acidity versus atmosphere-derived sources.

Many of the catchment-based processes of importance to surface water chemistry can undergo substantial changes over time, as the result of both natural and anthropogenic (but local) events. Perhaps most important in eastern Canada are the processes of forest aggradation and removal. Several researchers have shown that changes in forest vegetation can have important hydrological implications (Jeffrey 1970; Swank and Douglas 1974; Mahendrappa 1982; Mahendrappa and Kingston 1982), especially as these changes affect evapotranspiration and soil moisture retention. Hydrological processes play a key role in virtually all models of surface water acidification.

The vegetation present also plays a more direct role in catchment water chemistry by acting as both a source and a sink for many of the cations and anions of importance (e.g., Likens et al. 1977). Decaying vegetation is the primary source of organic anions to the system, while an aggrading forest may provide an important sink for acid anions and base cations, thereby modifying the ANC of the catchment's soils. Therefore, changes over time in the vegetative structure of a catchment

could conceivably lead to changes in surface water chemistry that mimic acidification due to atmospheric deposition.

One of the key arguments offered by proponents of the notion that catchment-based processes are more important than atmospheric deposition is that the soils in sensitive catchments are naturally highly acidic (Rosenquist 1978; Krug and Frink 1983; Noggle et al. 1984). The exchangeable acidity of soils such as podzols far exceeds the amount of acidity deposited from the atmosphere and, therefore, the relatively small additions of acidity from the atmosphere cannot significantly affect runoff chemistry.

If catchment-based natural processes overwhelm acidic deposition in determining surface water chemistry, then, as stated in M. L. Jones et al. (1986, p. 22):

> one would generally expect to see the following:
>
> 1. the principal anions associated with runoff acidity would be organic, because this is the primary form of acidity (produced from soil processes); and
>
> 2. the amount of acidity (mineral or organic) entering runoff should be independent of the amount of acidic deposition occurring in the catchment in question, provided acidic deposition [does not have the same geographic distribution as land use patterns that affect surface water chemistry].

Although organic anions have been shown to contribute significantly to the acidity of some surface waters, many studies have shown that where mineral acid deposition is substantial, mineral anions tend to dominate the chemistry of runoff and surface waters. For example, Cronan et al. (1978) found sulfate to be the predominant anion in a New England forest soil, while in the Pacific Northwest, where deposition is much lower, bicarbonate and organic anions were dominant.

The relative importance of mineral and organic acids in central Ontario was examined by LaZerte and Dillon (1984). At Plastic Lake, a small oligotrophic lake in the Muskoka-Haliburton district of Ontario,

they conducted a detailed mass balance study to look at the relative importance of sulfate and organic anions as sources of acidity. They found that while organic acidity dominates the runoff in this system during summer low-flow periods, sulfate is far more important during high-flow periods and on an annual basis. Several other studies have documented similar seasonal patterns in the relative contributions of organic and mineral anions (Kerekes et al. 1982; Visser 1984; Clair 1985; Kerekes et al. 1986a,b; Kessel-Taylor 1986). Furthermore, hydrogen ion shows a similar temporal pattern to sulfate in these systems, although its concentration typically exceeds that of sulfate in these cases.

Gorham et al. (1986) analyzed the chemistry of 37 Nova Scotian lakes and ponds, performing correlations among the major ion, pH, ANC, and DOC data. Their results indicate that the acidity of these waters is affected by both organic acids generated in the watersheds and by acidic deposition from long-range and local sources. SO_4^{2-} is primarily supplied to these lakes from atmospheric deposition, with the result that SO_4^{2-} deposition has caused naturally acidified waters to become more acidic. The Gorham et al. (1986) study illustrates that a thorough understanding of the natural acid-base balance of surface waters is required before attempts can be made to estimate the chemical effects of acidic deposition.

The presence of organic anions in surface waters is not the only indicator of non-anthropogenic acidification. Addition to certain soils of a neutral salt of a mobile, strong-acid anion can result in H^+ and the strong-acid anion entering surface waters (Johnson, et al. 1985). Substantial inputs of marine salts occur in maritime regions of eastern North America; in these areas such an effect could be of some importance. Although no specific analyses of this potential natural source of acidity exist for Canadian maritime waters, a recent U.S. study (Sullivan et al., 1988) found little support for the importance of this phenomenon, at least as a long-term acidifying process on a regional scale, for northeastern United States lakes.

In summary, our current process-level understanding of the sources and causes of surface water acidification indicates that both atmospheric and catchment-based contributions can be important. However, in all cases cited above, where acidic deposition is considered to be relatively high (i.e., where precipitation pH is less than 5.0 or SO_4^{2-} loading is greater than 20 kg $ha^{-1}year^{-1}$), mineral acid anions (principally sulfate) derived from atmospheric deposition make the greatest contribution by far to surface water acidity. The process-level evidence thus supports the contention that acidic deposition is at least in part responsible for surface water acidification.

3.3.3 Direct Evidence

In most regions of northeastern North America, acidic deposition does not appear to have changed substantially over the past decade or so, although some evidence is available of local decreasing trends around Sudbury (Keller and Pitblado 1986; Sect. 6.2), in Nova Scotia (Thompson 1986; Sect. 6.2), and in New Hampshire (Hedin et al. 1987). Because accurate and reliable measurements of key chemical variables have only recently been possible, there is relatively little direct evidence of the response of aquatic systems to changes in deposition. The single exception to this is the Sudbury region of Ontario. Between the mid-1970s and the early 1980s, sulfur emissions in this area declined by approximately 54% (Dillon et al. 1986). Several lakes have been monitored in this area during this period, some relatively intensively (Dillon et al. 1986), and others synoptically (Keller and Pitblado 1986; Keller et al., 1986).

Dillon et al. (1986) assessed the chemistry of five acidified lakes in the Sudbury area, four of which have been monitored continuously since 1973. The fifth was sampled twice, first in 1977 and then in 1982 (LaZerte and Dillon 1984). Declines in lake sulfate levels were observed in all five lakes (range: 25% to 62%) during the

period that emissions declined. Three of the lakes were subjected to experimental chemical manipulations during this period, which also influenced their pH and ANC. The remaining two lakes, however, exhibited increases in pH of 0.38 (4.23 to 4.61) and 0.84 (3.96 to 4.80) during the period of sulfate decrease. Finally, aluminum concentrations declined in these two lakes by 56% and 75%, respectively. Aluminum concentrations are known to increase as surface waters acidify, and aluminum is thought to be an important source of toxicity to biota, at least in clearwater systems (Jones, M. L., et al. 1986; Turner et al. 1986).

During the period 1981 through 1983, Keller and Pitblado (1986) resampled 209 lakes in the vicinity of Sudbury that had been sampled previously during the period 1974 through 1976. On average, they observed significant decreases in lake sulfate and increases in pH. They also found that the magnitude of change between the two survey periods was associated with the distance from Sudbury, with lakes closer to the smelters exhibiting more pronounced improvements. These studies and the issue of lake recovery from acidification are discussed in Sect. 6.

Krug and Frink (1983) suggested that even if mineral acid deposition contributes significantly to the acidity of surface waters in areas receiving high deposition rates, reductions in deposition will simply lead to organic acidity replacing mineral acidity. As pointed out by LaZerte and Dillon (1984), the evidence refutes this argument. In both Clearwater and Swan Lakes, two of the lakes discussed by Dillon et al. (1986), the pH increased in response to decreased lake sulfate, and no significant change in DOC was observed. LaZerte and Dillon (1984) also argue that an unreasonable amount of organic acidity would be required [based on the Oliver et al. (1983) model of organic acids] to replace the mineral acidity currently being delivered to Plastic Lake and to its watershed.

3.3.4 Evidence from Regional Surveys

Evidence from regional surveys that acidic deposition is responsible for changes in surface water chemistry comes from two types of analyses:

1. comparisons of lake sulfate levels with sulfur deposition rates on a regional scale; and
2. comparisons of lake sulfate levels and lake DOC or organic anion levels with lake ANC (alkalinity) and base cations.

Thompson and Hutton (1982, 1985) considered the relationship between wet sulfur deposition and lake sulfate levels by first correcting the latter for runoff to obtain a measure of watershed sulfate yield and then plotting deposition and yield across an east-west geographical gradient. They found a reasonably good association between yield and wet deposition across a compilation of lake chemistry data sets, ranging from northwestern Ontario to Labrador. The greatest discrepancy between yield and wet deposition was found in areas where dry sulfur deposition was presumed to make a relatively large contribution to the total deposition (i.e., southern and northeastern Ontario, southern Quebec). This relationship between sulfate in deposition and in runoff precludes the existence of significant watershed sources of sulfur (e.g., bedrock). They concluded that "the good agreement between sulfate yields and total deposition data indicates that there is little retention of sulfate within the watersheds or in the lakes, and that most, if not all, of the sulfate in the lakes can be ascribed to atmospheric deposition" (Thompson and Hutton 1985, p. 82).

Kelso et al. (1986a) also looked at the relationship between sulfate yields and assumed regional sulfate deposition levels (their Table 6). They found a significant, positive relationship ($p < 0.05$, $n = 806$, $r^2 = 0.43$) between these two variables. Jeffries et al. (1986) also note this pattern for the lakes included in their data set, as do Talbot et al. (1984) for lakes in Quebec.

Detailed work in smaller geographic areas has shown that lake water sulfate concentration may vary considerably, even though atmospheric deposition varies little. In Algonquin Park, the lake concentrations of sulfate vary by a factor of 2 (P. J. Dillon, Ontario Ministry of the Environment, personal communication to R. B. Cook, 1987). In northern Minnesota, north-central Wisconsin, and the Upper Peninsula of Michigan, lake water sulfate concentrations vary by a factor of up to 4, even though atmospheric deposition in the upper Midwest varies by a factor of 2 (Nichols and McRoberts 1986; Cook et al. 1987). The cause of the local variation in sulfate is sulfate retention, probably by in-lake bacterial sulfate reduction (Kelly et al. 1987; Cook et al. 1987). Sulfate retention is more important in mid-continental areas than in eastern North America, because lakes in the mid-continent typically have longer water residence times, which allows in-lake processes to have a greater influence on lake water chemistry (Kelly et al. 1987).

Jeffries et al. (1986) also considered the relationship between lake sulfate and ANC on a broad regional scale. They did not find a statistically significant relationship, although there was some indication that in areas receiving relatively high deposition, low ANC lakes were more likely to be associated with high lake sulfate levels than they would be in areas experiencing less deposition. They maintained that this result was not at all surprising, in view of the fact that biogeochemical factors that vary substantially on a local scale profoundly influence the interaction between acidic deposition and lake ANC. In other words, local, within-catchment variations in bedrock, soils, and vegetation combine with regional variations in atmospheric chemistry to determine surface water chemistry so that, for lakes within a region, water chemistry will exhibit a range of values.

Jeffries et al. (1986) also considered the relationship between ANC and organic anion concentrations (A^-). They found very little variability among regions in A^- except in northwestern Ontario, where levels were particularly high. ANC and A^- distributions appeared

to be completely unrelated to each other. They argued that the lack of a relationship between A^- and ANC on a regional scale strongly disputes the notion of a primary role for organic acidity in determining the current status of lakes in eastern Canada.

3.4 REGIONAL EVIDENCE FOR MAGNITUDE OF CHANGE

In the preceding section we presented evidence that supports the hypothesis of a causal relationship between acidic deposition and surface water chemistry. If one accepts the simple view of surface water acidification first proposed by Henriksen (1979, 1980), then one can use the current chemistry of lakes to draw inferences about the magnitude of change that has occurred as a result of acidification. Analyses of this sort have been presented in earlier assessments (e.g., Harvey et al. 1981; MOI 1983) and will not be repeated here.

More recently, Wright (1983) examined 27 lake chemistry data sets from Europe and North America. Based on the assumption that the chemistry of surface waters in areas remote from sources of acidic deposition indicates the original (preacidification) chemistry of lakes in less remote areas, he showed that the sulfate levels in the latter had risen considerably over historical levels. He also showed that much of this increase in sulfate levels was apparently concomitant with a decrease in the lakes' ANC (or increasing acidity), while a smaller proportion was matched by an increase in the lakes' base cation concentrations.

Johnson (1985) applied the Wright-Henriksen models to a series of 19 rivers draining into Georgian Bay in Ontario. He estimated an average ANC decline (original-to-present) of 107 to 147 $\mu eq\ L^{-1}$ and average increases in sulfate of 108 to 125 $\mu eq\ L^{-1}$, averaged over all 19 rivers, for various minor variations in the basic model. Kelso et al. (1986a) applied Henriksen's nomogram to the lakes in the NIS data set and found, respectively, 1.6, 1.4, 47.3, 15.4, and 4.0 percent of the lakes to be acidified according to Henriksen's criteria for Ontario, Quebec, Nova Scotia, New Brunswick, and Newfoundland (see Kelso et al. 1986a, Table 9).

Ion ratios can also be used to estimate the magnitude of change due to acidification, based on exactly the same assumptions that apply to the Wright-Henriksen models. Kelso et al. (1986a) examined the relationship between ANC and (Ca + Mg) for lakes in the NIS data set and found ANC deficits [i.e., ANCs of less than (Ca + Mg) in equivalent units] to be most pronounced in areas receiving the highest deposition levels.

Jeffries (1986) considered the ratios of ANC to (Ca + Mg), sulfate to (Ca + Mg), and ANC to sulfate for the overall Canadian data base described in Sect. 2.2. The regional patterns of these ratios correspond closely to patterns of atmospheric sulfur deposition (Fig. 3.2). The ANC:(Ca + Mg) ratios (Fig. 3.3) are lowest in south-central Ontario, New Brunswick, and Nova Scotia, slightly higher in Quebec and Newfoundland, and highest in northern Ontario and Labrador. Consistently high values of ANC:(Ca + Mg) are found in other areas that have low-ANC surface waters yet receive low rates of atmospheric deposition, for example in the western United States (Melack et al. 1985; Landers et al. 1987), in addition to northwestern Ontario and Labrador. This difference in ratios for areas having low as opposed to high atmospheric deposition suggests that low values may be due to atmospheric deposition.

Low values of ANC:(Ca + Mg) may also possibly result from surface waters that contain large concentrations of sodium and potassium (e.g., those waters near the ocean), appreciable NO_3^-, or large concentrations of organic anions. (Ca + Mg) are the primary base cations in surface waters because they are the cations that are most easily weathered from the types of bedrock and soils in eastern Canada (Stumm and Morgan 1981). The high values of Na in surface waters near the ocean are due to sea-salt aerosols; their effect can be removed by considering the non-sea-salt fraction. In areas remote from the influence of sea-salt aerosols, (Ca + Mg) are the primary base cations. NO_3^- concentrations are typically low in the lake data considered in Sect. 2, and NO_3^- does not contribute significantly to the total anionic charge. Organic anion concentrations are not related to ANC and show little variation in eastern Canada, except for northwestern Ontario (Jeffries et al. 1986).

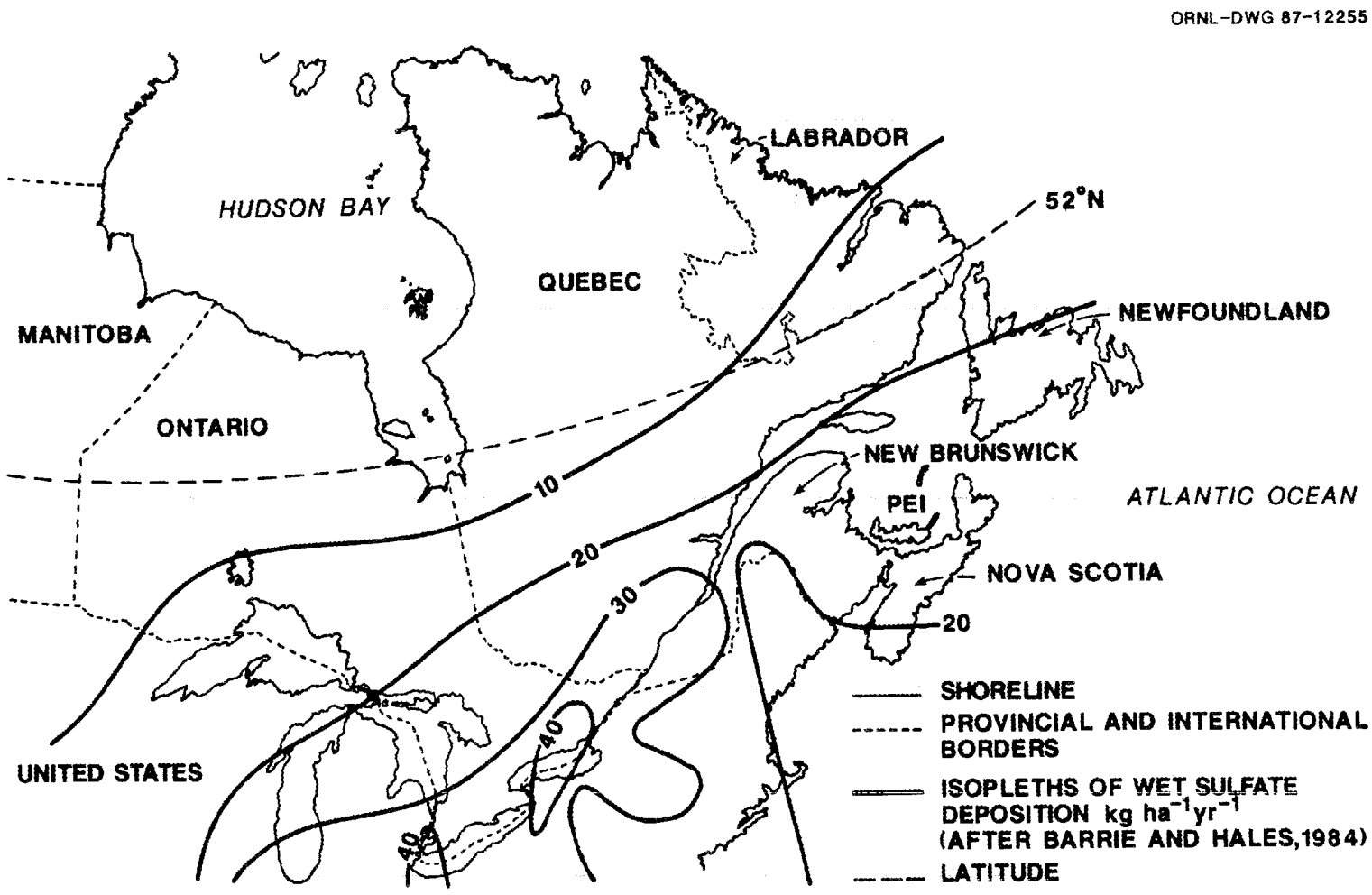

Fig. 3.2. The approximate isopleths of wet sulfate deposition in 1980 (after Barry and Hales 1984). Source: Minns and Kelso (1986).

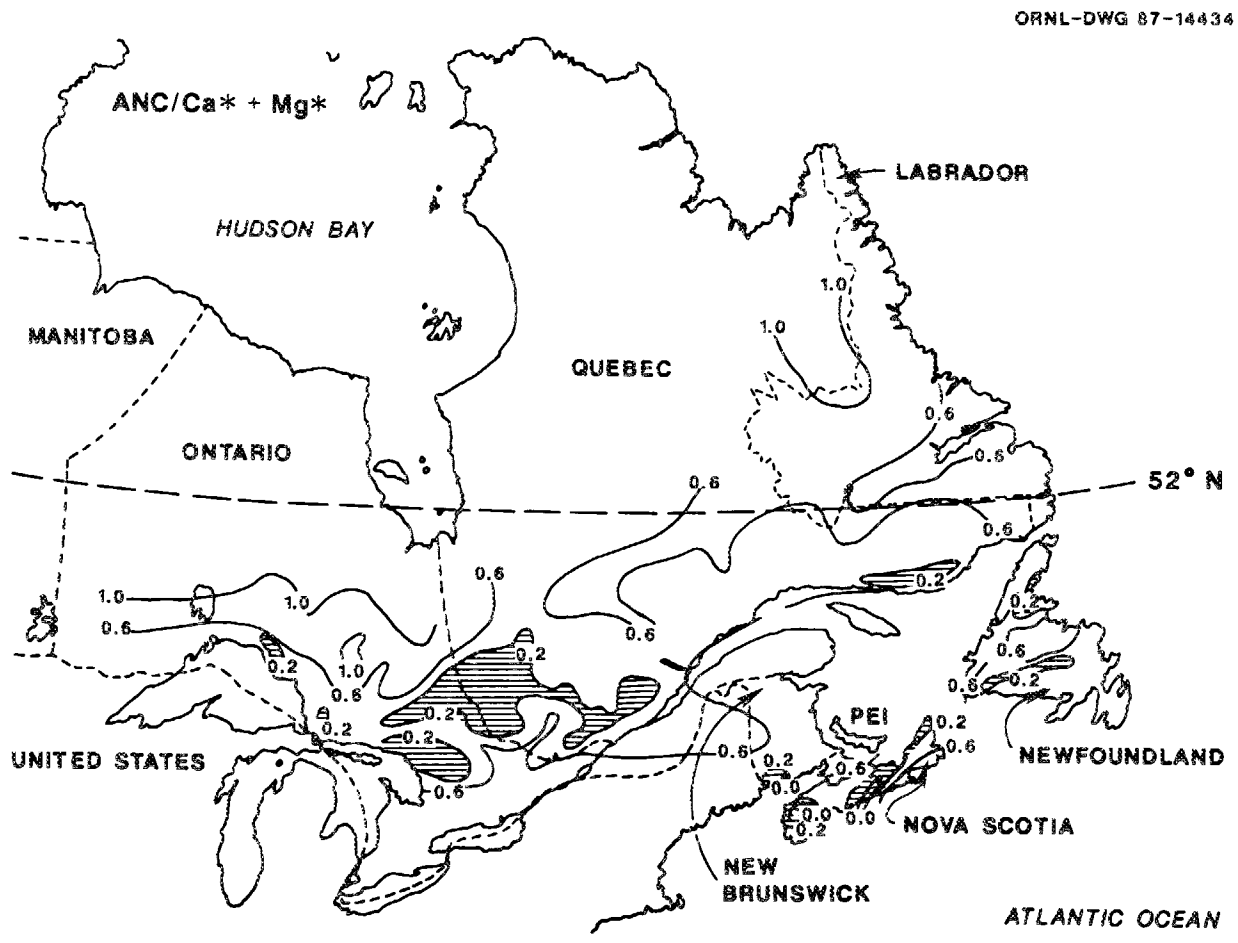

Fig. 3.3. Geographical distribution of ANC:(Ca* + Mg*) in eastern Canada. Asterisks signify that Ca and Mg have been corrected for sea salts. Low values of ANC:(Ca* + Mg*), indicated by cross hatching, are found in areas of relatively high sulfate deposition, suggesting that the low values are due to atmospheric deposition. Source: Jeffries (1986).

Low values of ANC:(Ca + Mg) may also result from increased weathering of base cations. For certain terrains, high SO_4^{2-} deposition may cause an increase in base-cation weathering rather than a decrease in ANC. However, for most areas of the Precambrian Shield in eastern Canada, the increase in base-cation concentration is only 0 to 40% of the increase in sulfate deposition, with the majority of the increase in sulfate deposition causing a decrease in ANC (see Sect. 5).

Sulfate:(Ca + Mg) ratios (Fig. 3.4) are high when ANC:(Ca + Mg) ratios are low, as could be expected if ANC is being replaced by sulfate. Finally, ANC:sulfate ratios (Fig. 3.5) are similar to ANC:(Ca + Mg) ratios, except that the former are considerably higher in remote areas. Taken together, the three ratios show similar geographical distributions, which are consistent with SO_4^{2-} from atmospheric deposition replacing lake water ANC in areas receiving high levels of acidic deposition.

Analyses of regional survey data, therefore, generally indicate that a substantial acidification has occurred in areas of eastern Canada receiving elevated levels of acidic deposition. It must be emphasized, however, that this interpretation of the survey data is based on key assumptions regarding both the basic processes involved in surface water acidification and the validity of using data from one region as being indicative of the historical chemical status of lakes in another region. Nevertheless, the regional distribution of lake sulfate levels does appear to correspond well to deposition. Furthermore, both regional surveys and detailed studies suggest that alternative sources of acidity are not likely to be of great importance, either on a regional scale or in areas receiving substantial acidity from atmospheric sources. Finally, where acidic deposition has decreased appreciably in recent years, there is good evidence that lakes respond with a decrease in SO_4^{2-} and an increase in pH and ANC.

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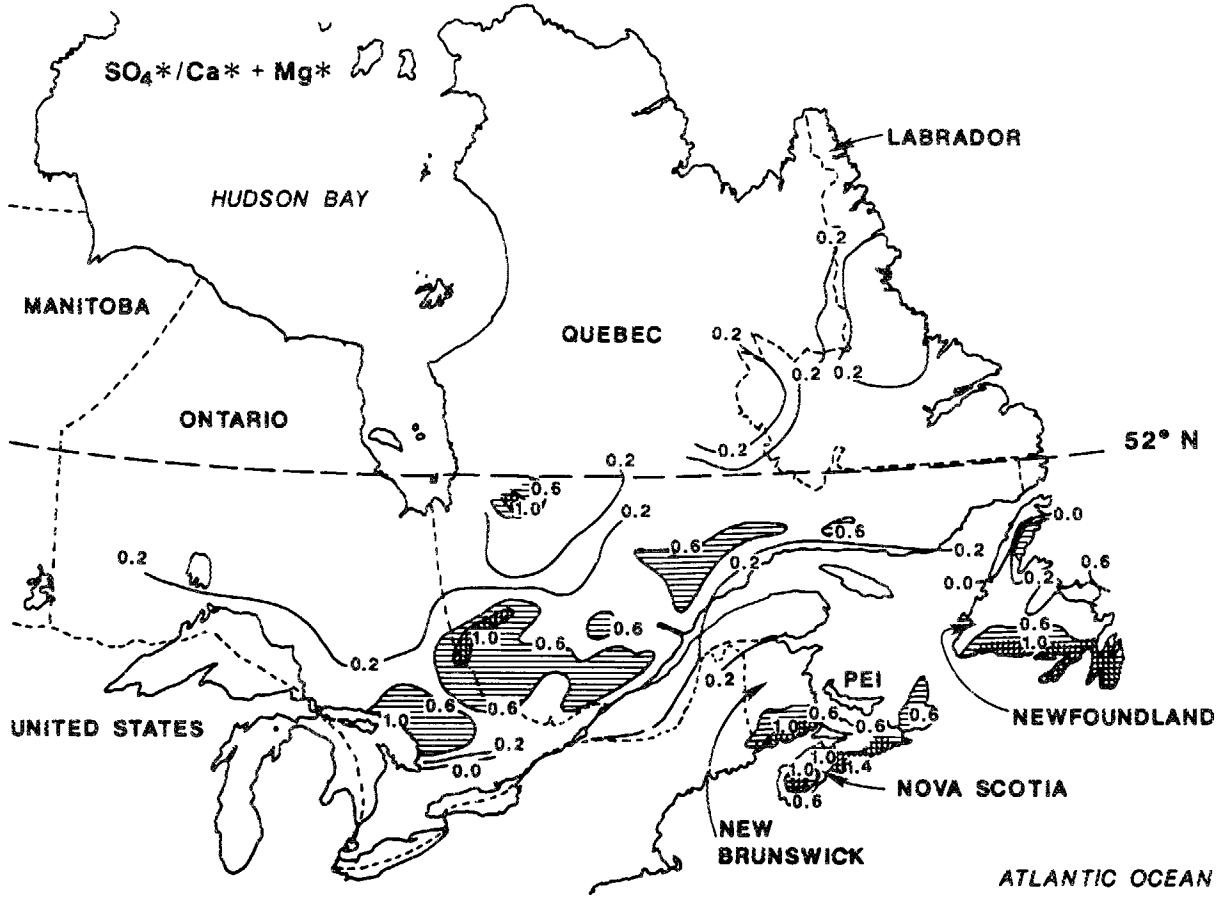

Fig. 3.4. Geographical distribution of SO_4^{2-*}:(Ca* + Mg*) in eastern Canada. Asterisks signify correction for sea salts. High values of SO_4^{2-*}:(Ca* + Mg*), indicated by cross hatching, are found in areas of relatively high sulfate deposition. Source: Jeffries (1986).

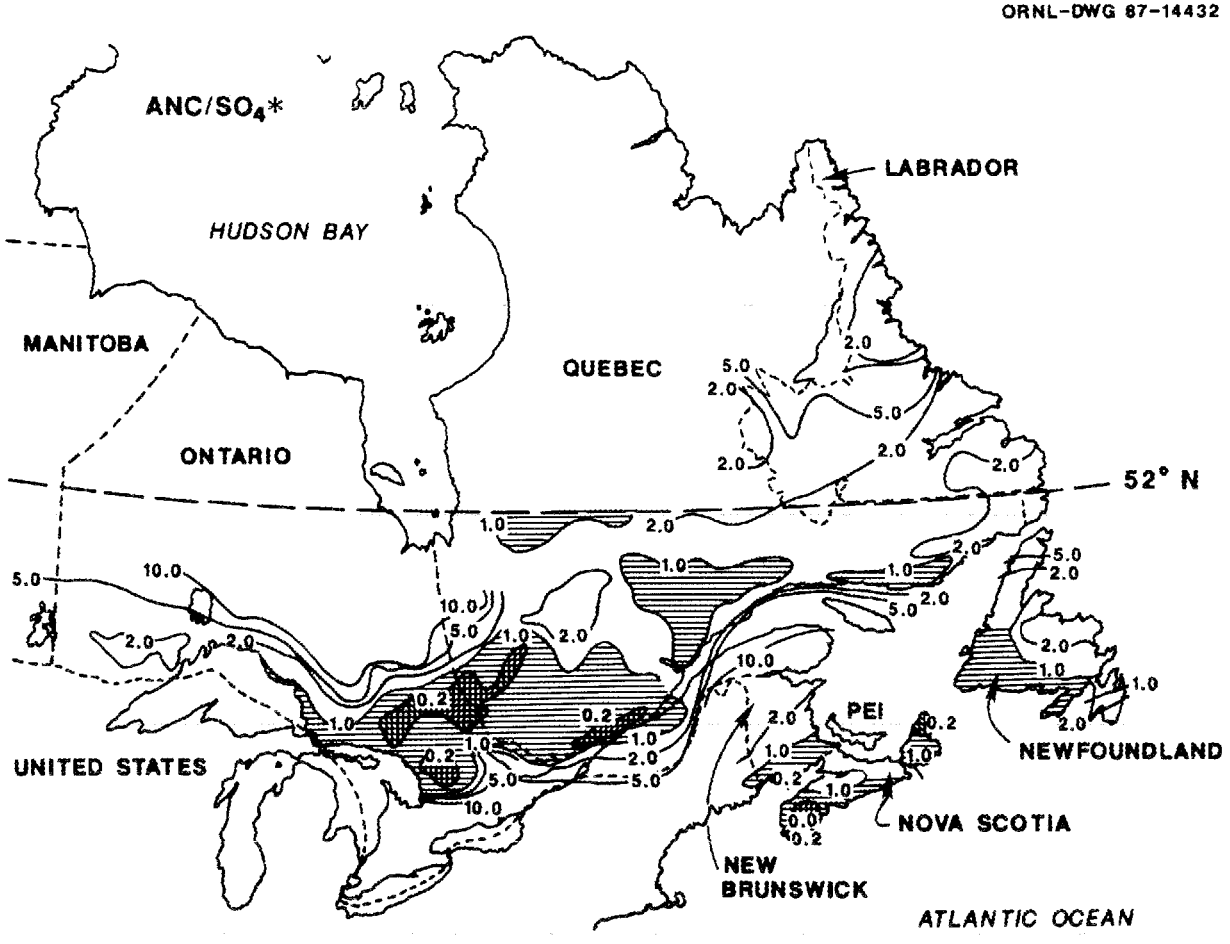

Fig. 3.5. Geographical distribution of ANC:SO_4^{2-*} in eastern Canada. Low values of ANC:SO_4^{2-*}, indicated by cross hatching, are found in areas of relatively high sulfate deposition, suggesting that the low values are due to atmospheric deposition. Source: Jeffries (1986).

3.5 SUMMARY

Paleolimnological data for eastern Canada are insufficient to facilitate examination of lake acidification caused by the long-range transport of acidic deposition. Of the five lakes for which pH reconstructions using diatom or chrysophyte remains have been published, none indicated acidification. However, four of the lakes have relatively high ANCs and would not be expected to acidify from atmospheric deposition. Preliminary results from diatom and chrysophyte studies in 12 Quebec lakes and chrysophyte studies in 6 Ontario lakes indicate that these presently acidic lakes have undergone a fossil-inferred pH decline in recent years. The limited evidence from sediment chemistry profiles of lakes in Canada remote from point sources indicates a history of changing atmospheric chemistry over the past 100 years. Sediment chemistry data cannot be used to estimate acidification, however.

Historical surface water chemistry data for the past 10 to 50 years are limited. Where data exist, changes in pH and ANC are consistent with acidic-deposition-caused changes in surface water chemistry, although the pH trends are obscured by measurement uncertainties of historical data. In regions where paleoecological techniques have been used in conjunction with an analysis of historical data (e.g., southern Ontario), results from both techniques supported a conclusion that surface waters have been affected by atmospheric deposition.

Processes occurring within the soils, vegetation, and surface waters of a catchment may greatly influence the chemistry of surface water, both by mediating the effect of acidic deposition on water chemistry and by providing sources of acidity. Several studies have shown that organic acids, presumably produced in the watershed, can contribute significantly to the acidity of surface waters. However, many studies have shown that in areas where mineral acid deposition is substantial, mineral anions tend to dominate the chemistry of runoff and surface waters.

There is relatively little direct evidence of the response of aquatic systems to changes in deposition. Emissions in the Sudbury region of Ontario declined by 54% between the mid-1970s and early 1980s, and decreases in lake sulfate concentrations and increases in pH and ANC were observed. Thus, where acidic deposition has changed appreciably in recent years, the SO_4^{2-}, pH, and ANC of lake water respond as expected.

Evidence from regional surveys shows that acidic deposition is largely responsible for the acid-base chemistry of surface waters. Comparison of sulfate deposition and lake water sulfate reveal that most of the sulfate concentration variation of lakes can be ascribed to atmospheric deposition. Analyses of ion ratios indicate that ANC has been replaced by SO_4^{2-} over broad areas of eastern Canada, resulting in greatly decreased ratios of ANC:(Ca + Mg) and increased ratios of SO_4^{2-}:(Ca + Mg). Furthermore, analyses of the relationship between organic anions and ANC on a regional scale strongly disputes the notion of a primary role for organic acidity in determining the current status of lakes in eastern Canada. The regional patterns of ion ratios correspond closely to patterns of atmospheric sulfur deposition, indicating a much greater extent of acidification in areas receiving high levels of acidic deposition.

4. BIOLOGICAL RELEVANCE OF OBSERVED CHANGES IN CHEMISTRY RESULTING FROM ACIDIC DEPOSITION

(Jerry W. Elwood)

As its primary focus, this section addresses the following question: What evidence is there to support the hypothesis that biological changes in surface waters in Canada resulted from chemical changes associated with acidic deposition? The objective is to assess the evidence concerning the causal relationship between biological changes and water-quality changes associated either directly or indirectly with acidic deposition.

4.1 CONCEPTUAL FRAMEWORK OF AQUATIC EFFECTS

Aquatic species differ in their sensitivity to pH, as shown by numerous studies of fishes, zooplankton, phytoplankton, benthic invertebrates, and decomposers (Magnuson et al. 1984). Most interest is focused on the impacts of acidic deposition on fishes because of their clear economic and sociological importance. In addition, fishes depend on lower trophic levels and, thus, serve as integrators of ecosystem function. Therefore, in the assessment of the biological status of Canadian surface waters, fish are the main, but not the only, consideration. In Sect. 4.2, the current status and historical trends among aquatic biota in Canadian surface waters are assessed in terms of their response to acidification.

The evidence used to assess biological changes in streams, rivers, and lakes is based on both direct and indirect empirical data. Direct evidence includes (1) data on long-term changes in biological communities and populations in surface waters that have exhibited changes in water chemistry over the same time period, (2) synoptic biological surveys of surface waters that have a range in pH acidification, and (3) results of in situ bioassays in which the survivorship, physiological status, or behavior of organisms and communities in more acidic surface waters are compared with those of the same species or community in a control or reference environment that is relatively less acidic. Indirect empirical evidence includes

both data on biological changes in surface waters that are experimentally acidified and results of laboratory bioassays that provide data on threshold concentrations at which biological effects on populations and communities in natural environments would be predicted.

4.2 LONG-TERM TRENDS IN AQUATIC BIOTA

Analyses of time-trends in the biological status of surface waters rely on comparing historical and current field survey data. Such observations of trends can verify that species are actually being lost. By themselves, however, they can not be used to estimate the extent and magnitude of impact. Furthermore, observed biological changes through time, with or without associated evidence of increasing acidity, do not prove that acidification is the cause, nor do they disprove alternative explanations of observed trends.

Trend analyses of biological changes are best documented in two areas of Canada: the La Cloche Mountain lakes (173 total) located near Sudbury, Ontario, on the north shore of Georgian Bay and the North Channel of Lake Huron, and Atlantic salmon rivers in Nova Scotia. Research on 68 of the largest La Cloche Mountain lakes began in the 1960s with periodic surveys that provided a data base for time-trend analysis. Historical records of sports fish were compiled from reports from anglers and conservation officers, as well as from limited surveys (Harvey and Lee 1982). Results indicate the probable loss of lake trout (*Salvelinus namaycush*) from at least 17 lakes, loss of smallmouth bass (*Micropterus dolomieu*) from 12 lakes, and loss of walleye pike (*Stizostedion vitreum*) from four lakes. Both yellow perch (*Perca flavescens*) and rock bass (*Ambloplites rupestris*) are reported as being lost from only two lakes. Although there were no historical reports of fish captures for many of the La Cloche lakes, the documented lost populations are probably underestimates of total losses, because many fish species that are not currently present in some lakes are likely to have had a wider distribution historically than is indicated by current surveys.

The exact cause of these losses of fish species from La Cloche Mountain lakes is less certain. Because both sport and nonsport fishes declined, overfishing is unlikely to have been solely responsible for the decline (Beamish and Harvey 1972). Acidification was hypothesized to be the cause of the fish extinctions because many of the lakes in and to the east of the La Cloche Mountains had a low pH in 1971 and had experienced a decline in pH over the preceding 3- to 13-year period (Beamish and Harvey 1972). The pH measurements taken in the early 1960s were made using the colorimetric method, and thus the reported values might be too high. If so, the reported reductions in pH for many of the La Cloche Mountain lakes would be in error. However, the magnitude of pH reductions in La Cloche Mountain lakes (up to 2 pH units) is much greater than the error attributable to differences in measurement errors [<0.2 pH units in general and <0.5 units in the worst case (Kramer et al. 1986)].

Harvey (1975) used regression analysis in an attempt to identify the causal factor responsible for variation in the number and diversity of fish species among lakes and, hence, to indirectly identify the potential cause of the loss of fish species from lakes. The number of fish species in the La Cloche lakes in 1972 and 1973 was significantly correlated with pH ($p < 0.005$), although the correlation accounted for only 39% of the variation among lakes in the number of fish species present (Harvey 1975). The number of fish species present was also significantly correlated with other physical, chemical, and biological variables, including lake depth, volume, Secchi depth, hardness, and number of zooplankton species (Harvey 1975). Baker (1984) performed an analysis of covariance on the La Cloche data and found that the correlation of number of fish species with pH was significant ($p < 0.005$) even after adjustment for differences in lake areas.

The La Cloche Mountain lake data were also reanalyzed by Henderson (1984) to identify the environmental variables that constrain the fish community in these lakes. The frequency distribution of environmental variables--including pH, ANC, Ca, lake size, lake depth, and the presence of inlet or outlet streams--was plotted, after which the number of lakes in each class of variables with and without fish was determined. The effect of each variable on species distributions and

the interaction between the variables were then tested statistically. The variables found to significantly affect species distribution were then examined to identify the so-called "constraint" values outside of which a particular species of fish did not occur. Because little information was available on rare fish species present in the La Cloche lakes, the analysis was limited to the 14 species present in >10% of the lakes.

Factors found to influence the occurrence of these 14 species were calcium concentration, pH, lake area, the presence of streams connecting them to other lakes, and lake depth. The analysis indicated that pH and lake area were the two dominant factors determining the species composition in the La Cloche Mountain lakes. Of the 14 species considered in the analysis, three species (Iowa darter, Johnnie darter, and bluntnose minnow) were estimated to be constrained by pH alone, and three species (white sucker, rock bass, and smallmouth bass) were constrained by a combination of pH and lake morphology. Four other species are apparently constrained by calcium concentration. The effect of changing pH and lake area on the complement of fish species was predicted for the La Cloche lakes with a calcium concentration >5 ppm and a maximum depth >5 m (Fig. 4.1; also Fig. 1 in Henderson 1984).

While Henderson's (1984) analysis shows that a number of factors limit the species composition of fish in the La Cloche lakes and that these factors are different for various species, there is a significant correlation between water pH and the number of fish species present in these lakes. This result is thus consistent with the hypothesis that acidification has contributed to the loss of fish species from the La Cloche Mountain lakes.

As is to be expected, correlative techniques do not account for all of the variation in the number of fish species present in the La Cloche Mountain lakes, because many variables that could influence fish presence were not measured (e.g., trace metals, fishing pressure, and management practices). Nevertheless, although the precise contribution of these other factors to the decline in fish species is unknown, the strong association between pH and the number of fish species has been clearly demonstrated.

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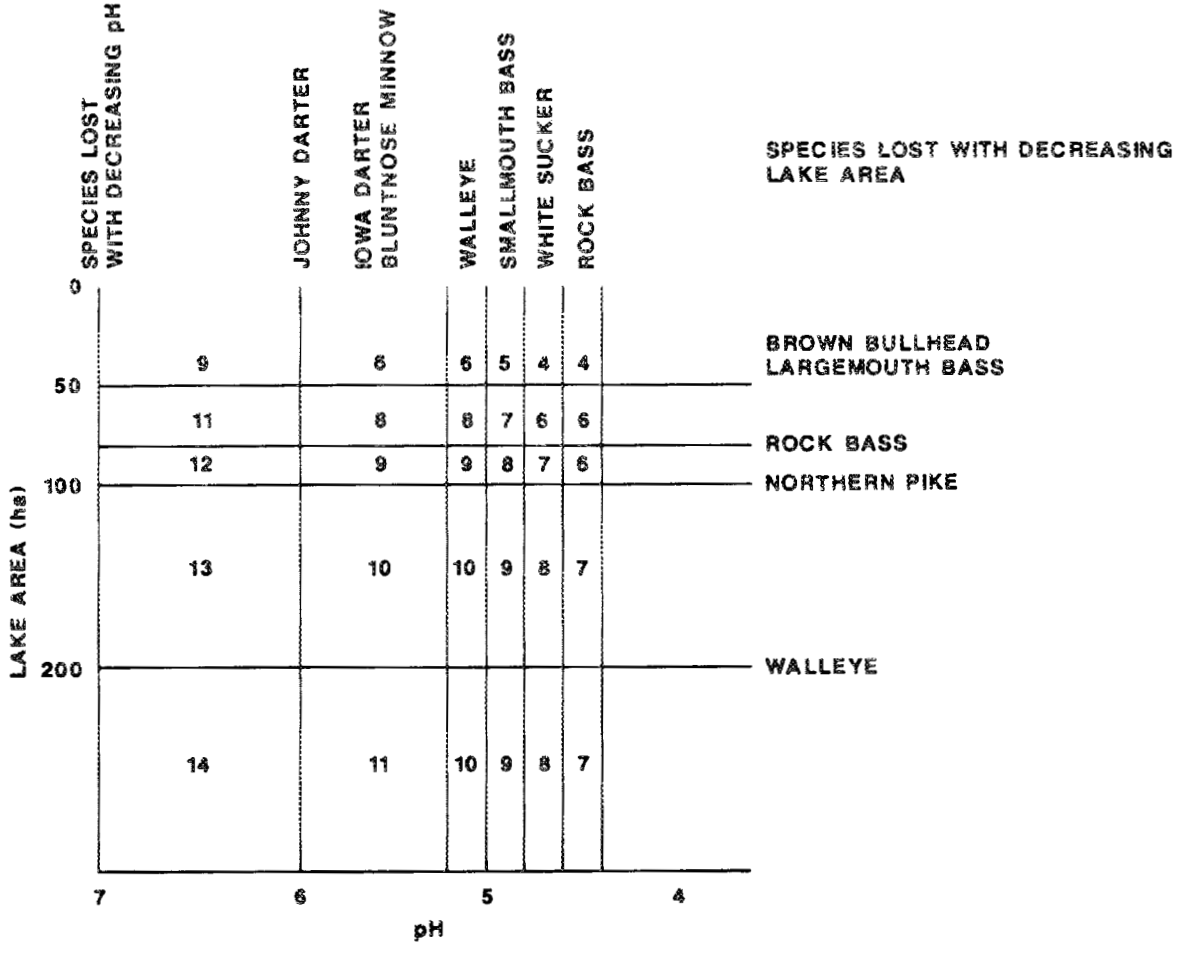

Fig. 4.1. Predicted effect of changes in lake area and pH on the number of fish species in La Cloche lakes. The predictions are for lakes with a calcium concentration >250 μeq L^{-1} and a maximum depth >5 m. Source: Henderson (1984).

As previously indicated, these known lost populations are probably underestimates of total losses because (1) the historical records of the presence of sport fish in lakes in this area are meager and (2) many of the sport fish species are likely to have had a wider distribution than is reflected in the results of recent surveys in the La Cloche Mountains (Harvey and Lee 1982). In addition, some sport fish are of less interest and, thus, their presence may not have been reported in the historical records.

To estimate the number of lost species of fish from the La Cloche Mountain lakes as a function of lake pH, Harvey and Lee (1982) compared the observed number of species in the late 1970s with the expected number as computed from an empirical relationship between number of fish species and area of lakes with a pH >6, in which acidification would not be expected to eliminate any species. Their results (Fig. 4.2; also Fig. 9 in Harvey and Lee 1982) show that below pH 5.2, 20% to 100% of the fish species are lost from the La Cloche Mountain lakes. Variation in the fraction of fish species lost as a function of pH is probably due to a combination of factors, including species differences in acid tolerance; existence and extent of refuges from acidic conditions within lakes; variation among lakes in the toxicity of metals mobilized via acidification; and differences among fish species and lakes in behavioral ecology, such as season and location of spawning.

Long-term records of water quality and angler success over a 27-year period are also available for Atlantic salmon rivers in Nova Scotia (Watt et al. 1983). These records show a significant decline in pH at an average annual rate of 0.017 pH units. Concentrations of bicarbonate also declined, while sulfate and aluminum concentrations increased over the same period. Atlantic salmon no longer run in rivers whose current pH is <4.7. In rivers whose current pH ranges from 4.7 to 5.0, salmon angling returns have declined significantly (Fig. 4.3; also Fig. 4 in Watt et al. 1983), and these rivers have low densities of juvenile salmon. Rivers having a pH >5.0 generally have normal densities of juvenile salmon and no trend in angler returns of Atlantic salmon (Fig. 4.3).

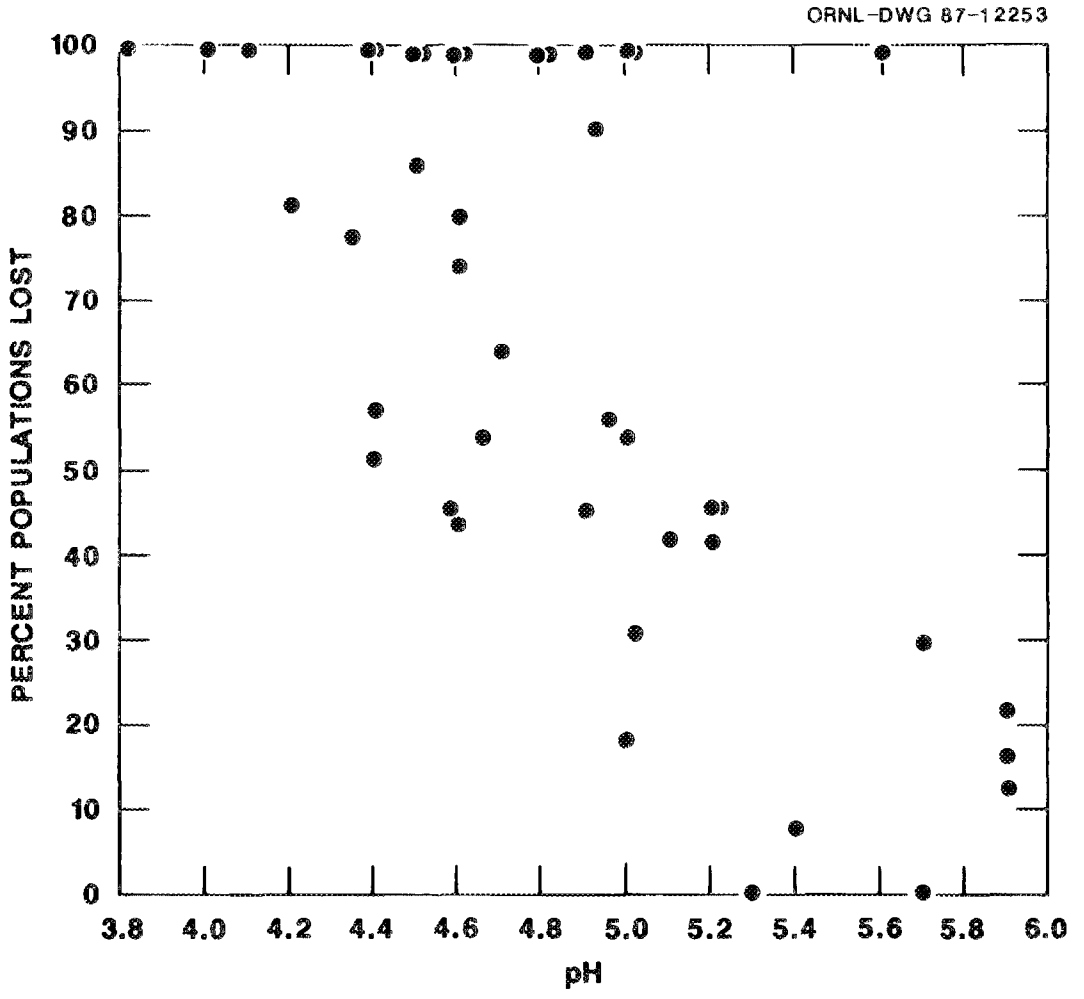

Fig. 4.2 Calculated percentage of the fish species lost from 50 lakes with pH <6.0 in the La Cloche Mountains as a function of pH. The lost populations were calculated as a fraction of the expected number of species in a given lake estimated from a regression model of lake area and number of fish species. Source: Harvey and Lee (1982).

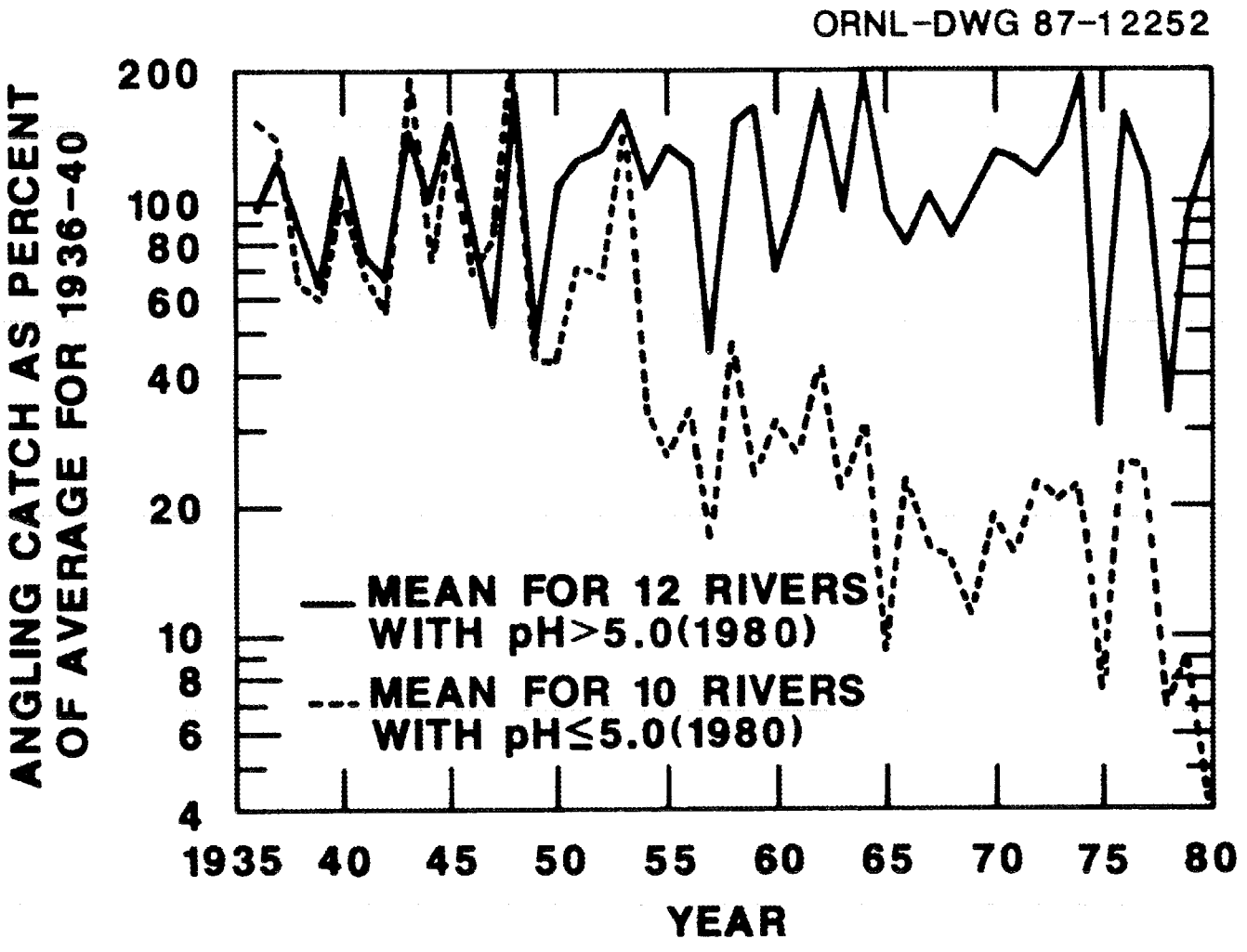

Fig. 4.3. Angler catch of Atlantic salmon, expressed as a percentage of the catch during the period 1936-40, versus time in years, in 12 Nova Scotia rivers having a mean pH of >5 and in 10 Nova Scotia rivers with a mean pH ≤5. Source: Watt et al. (1983).

Although the changes in pH, ANC, and sulfate in these Nova Scotian rivers are consistent with acidification-caused changes in surface water chemistry, they alone do not prove that this is the causal factor in the decline of salmon angling returns. The role of acidification, however, is corroborated by evidence of the effect of low pH on Atlantic salmon in laboratory bioassays (Daye 1980; Peterson 1984), in situ bioassays (Lacroix 1985; Lacroix et al. 1985), and electrofishing surveys (Watt et al. 1983). Taken together, these results generally support the conclusion that acidification contributed to the decline and loss of some fish species from these waters.

Whether the decline and apparent loss of Atlantic salmon from some rivers in southwest Nova Scotia can be attributed entirely to surface water acidification resulting from acidic deposition is open to question. Kessel-Taylor (1986) concluded that local factors, including changes in salmon management (e.g., over harvesting, habitat degradation) and land management practices (e.g., logging, agriculture), droughts, and the occurrence of natural sources of organic acidity produced in wetlands in the watersheds, have also had a significant impact on surface water acidity and salmon populations. Unfortunately, it is presently not possible to quantitatively determine the importance of each of these environmental factors on water quality and salmon populations, relative to that of acidic deposition. The data suggest that the cumulative effect of the above-mentioned factors, together with acidic deposition, contributed to the disappearance or decline of salmon from some rivers in southwest Nova Scotia.

4.3 SYNOPTIC SURVEYS

Synoptic surveys have been used to determine the current biological status of selected taxonomic groups in waters that have ANCs and pHs that could have been altered by acidic deposition and in waters considered potentially susceptible to acidic deposition. In selected areas or regions of eastern Canada, results of such surveys have

provided some initial qualitative indications of the current biological status of specific taxonomic groups (e.g., fish and benthic macroinvertebrates).

Kelso et al. (1986a) report the results of a fish survey conducted in 510 lakes in eastern Canada. The results of this survey, however, are confounded by differences in the sampling efforts of the different provinces (e.g., size of gill net set) and by biases in the types of lakes that were sampled. In Ontario and Newfoundland, for example, almost all lakes sampled were small headwater lakes, whereas in Quebec and Labrador, selection was biased toward larger lakes. Because lake size has been shown to influence the number of fish species (Harvey and Lee 1982), the influence of acidification on the number of fish species is potentially confounded by the effect of lake size. Kelso et al. (1986a), however, found that pH was not significantly correlated to the size of lakes in eastern Canada. Based on this finding and the trend of decreasing fish species with declining pH (Fig. 4.4; also Fig. 10 in Kelso et al. 1986a), they concluded that, of the parameters included in their survey, pH has the greatest influence on number of fish species. This trend alone, however, does not prove that acidity is the primary factor responsible for differences in the number of fish species in lakes in eastern Canada.

To determine whether the presence or absence of a fish species could be predicted based on the physical and chemical characteristics of a lake, Kelso et al. (1986a) performed a stepwise discriminant analysis of these lake survey data. Their results show that the discriminant functions were reliable in predicting the presence or absence of certain fish species in some areas but poor for other common species. In general, the distribution of brook trout, a species less sensitive to acidification (Grande et al. 1978; Baker 1984), did not depend on the water-quality factors related to acidification for which data were available (i.e., pH and ANC). In contrast, the distribution of lake trout and Atlantic salmon, which tend to be more acid-sensitive than brook trout, did depend on these factors. Therefore, these results

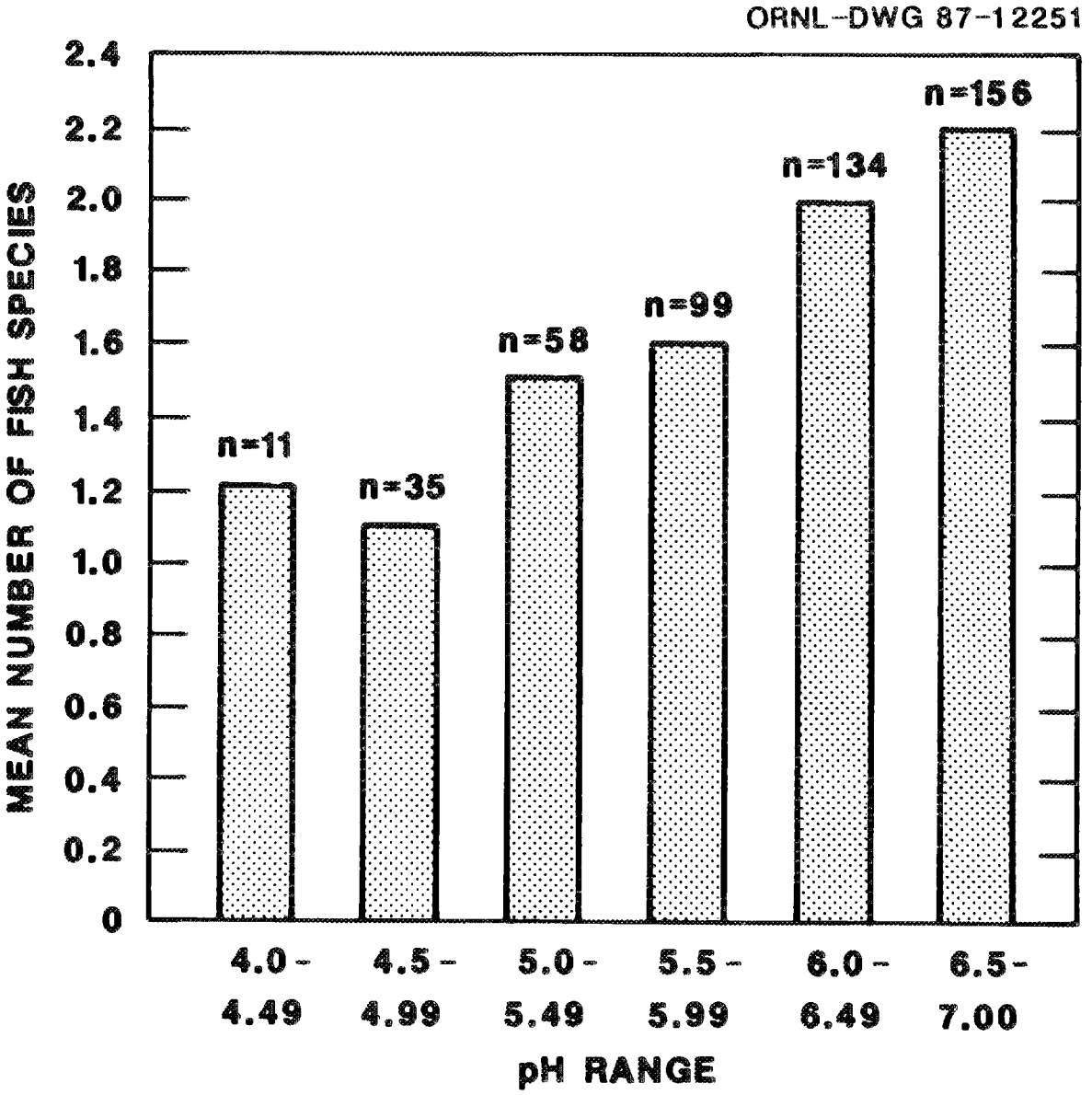

Fig. 4.4. Relationship between numbers of fish species and the lake pH for all lakes in eastern Canada. Source: Kelso et al. (1986a).

suggest that acidification has influenced the distribution of acid-sensitive fish species (e.g., lake trout and Atlantic salmon) in certain lakes in eastern Canada.

In the period 1982 to 1985, the presence, age distribution, and relative abundance of fish were determined in 36 lake trout (Salvelinus namaycush) lakes and 23 brook trout lakes in Ontario, spanning a range of water chemistry (Beggs et al. 1985; Beggs and Gunn 1986). All of the lakes surveyed were known to have contained lake trout or brook trout at one time, as documented in historical surveys and in reports from reliable conservation officers and anglers. Beggs et al. (1985) and Beggs and Gunn (1986) found that the relative abundance, as determined from catch per unit effort, of lake trout declined markedly in lakes with a pH <5.3 and total extinction occurred in lakes with a pH <5.2 [Fig. 4.5; also Fig. 6 in Beggs et al. (1985)]. Some of the fish lakes with extinct lake trout populations contained large populations of acid-tolerant species such as yellow perch (Perca flavescens) while lakes with a pH <4.7 were fishless. In lakes of pH 5.2-5.4, lake trout populations consisted almost entirely of large, old fish, indicating that successive recruitment failure was occurring at a pH of <5.5.

In contrast to lake trout, brook trout were more tolerant of low pH. They were captured at a minimum pH of 5.1 (Fig. 4.6, also Fig. 1b in Beggs and Gunn 1986), but populations were extinct below a pH of 5.0. There was no indication of reproductive failure of brook trout above a pH of 5.1.

This tolerance of brook trout to low pH is generally consistent with results of bioassay studies (Grande et al. 1978; Rosseland and Skogheim 1984) showing that brook trout are more tolerant of low pH than any of the other salmonid species tested. In addition, brook trout have the ability to detect and avoid acid water (Gunn and Noakes 1986), thus making them potentially less vulnerable to acidification if "adequate" (i.e., less acidic) refugia are available. Because brook trout locate their spawning sites in the presence of groundwater

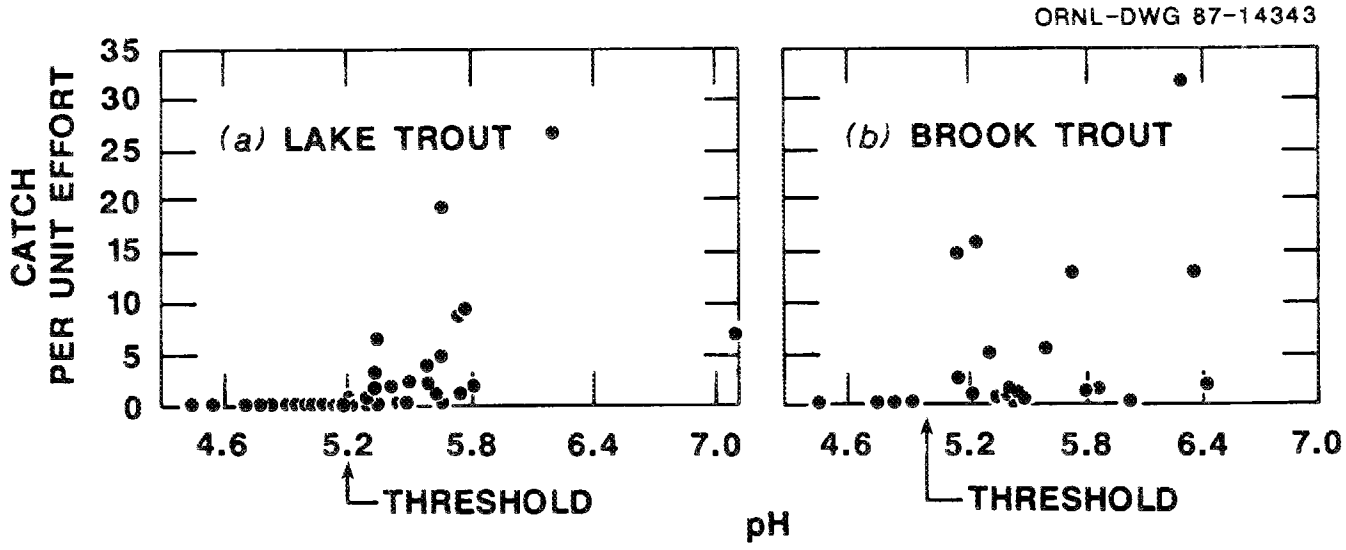

Fig. 4.5. Relationship between catch per unit effort of native lake and brook trout and midlake summer epilimnetic pH in 24 lakes in Ontario known to have contained lake trout in the past. (a) Values for native lake trout (Salvelinus namaycush); (b) values for native brook trout or char (Salvelinis fontinalis). The threshold value is the pH below which the catch per unit effort is 0 for these study lakes. Source: Beggs and Gunn (1986).

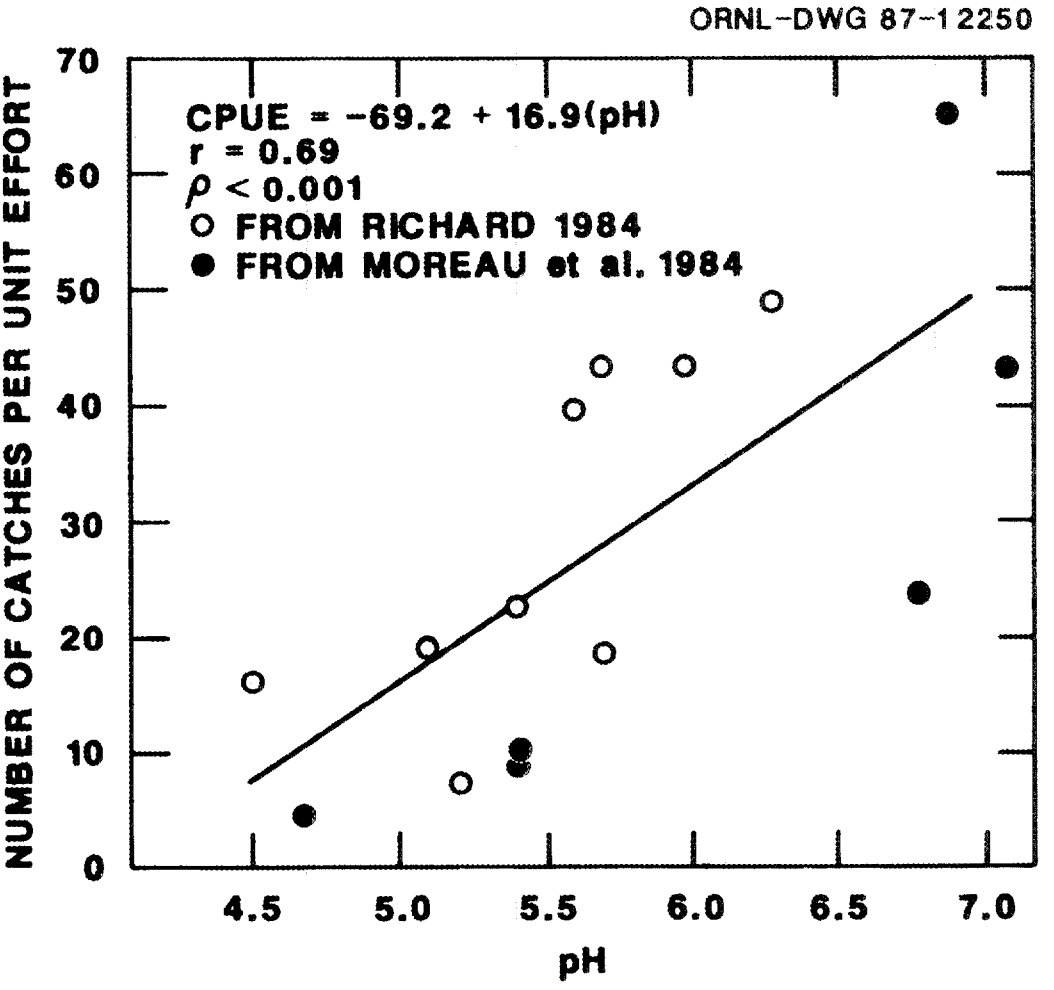

Fig. 4.6. Relationship between catch per unit effort of brook trout (Salvelinus fontinalis) in lakes in Quebec and pH of lake water. Source: Talbot et al. (1984).

upwelling that is generally of higher alkalinity than surface water (Gunn 1986), the early developmental stages (i.e., prior to emergence) of this species may be protected from acidic runoff and surface water.

The hypothesis that acidification caused the loss of lake trout populations from lakes with a pH <5.2 is supported by results from other surveys (Beamish 1976) and from the experimental acidification of a small lake (Schindler et al. 1985). Based on data from fisheries surveys of the La Cloche Mountain lakes, Beamish (1976) suggested that lake trout populations are lost in lakes having pH <5.2 to 5.5. This prediction was corroborated by Schindler et al. (1986) during the experimental acidification of a lake trout lake, Lake 223, in northwestern Ontario. Changes in fish reproduction and the condition of fish were followed over an 8-year period as Lake 223 was experimentally acidified from an original value of pH 6.8 to pH 5.0. Recruitment of lake trout ceased when the average pH of lake water was reduced to approximately 5.6, and no fish species reproduced at pH values of <5.4. Because lake trout are a long-lived species and have high natural survival rates, they will persist in a lake for many years after reproduction has ceased. On the basis of natural mortality rates of the most long-lived fish species, Schindler et al. (1985) predicted that the lake would become fishless within a decade if the pH remained below 5.4. The Beggs et al. (1985) observation that lake trout had become extirpated in lakes with a pH of <5.2 is thus consistent with these results and with the hypothesis that acidification was the cause of the loss of lake trout from lakes with a pH <5.2.

The interpretation that recruitment failure caused by acidic conditions occurs at a pH of <5.5 and is at least one of the causes for the extirpation of lake trout from some of the acidic lakes in Ontario is supported by time-series data on a lake trout lake that is recovering from acidification (Beggs and Gunn 1986). In 1978 and 1980 when the mean annual pH of Whitepine Lake near Sudbury, Ontario was 5.47, the lake trout population consisted of large, old individuals, indicating there was no recruitment. In 1982, however, when the mean pH of this lake had increased to 5.63, new recruits to the lake trout population were captured for the first time.

A stepwise discriminant function analysis was conducted by Beggs et al. (1985) to identify the physiochemical characteristics that differentiated the lakes having viable lake trout populations from those with extinct populations. Inorganic monomeric aluminum concentration alone--as calculated from an empirical relationship between pH, total Al, and DOC concentration in Canadian lakes--correctly classified the lake trout population status of 96.5% of the lakes for which the existing status of extinct and viable lake trout lakes was known.

The discriminant function using pH alone was then applied to a more extensive data set of assigned lake trout lakes in Ontario (1223 lakes) for which at least pH, alkalinity, and conductivity were known. Using this discriminant function, the number of lakes having either extinct or viable lake trout populations was estimated. Although the method used to assign a lake to the "lake trout" category is not described, it appears to have been based on both lake location relative to known glacial activity, the type of glacial deposit, and characteristics of the lake's watershed. This information on the 1223 lake trout lakes was then extrapolated to the entire lake trout resource in Ontario. According to this analysis, acidification has caused lake trout to become extinct in 3% (63 lakes) of the 2218 lakes in Ontario estimated to have lake trout. If the lakes around Sudbury are removed from this analysis because of the large influence of the point source emissions from the Sudbury smelters, only 1% of the lake trout lakes in Ontario would be acidic (ANC <0) and <1% of the non-Sudbury lakes would be estimated to have lost their lake trout populations as the result of acidification.

Synoptic surveys have also been conducted on approximately 200 lakes in four areas of Quebec since 1981 (Talbot et al. 1984). These various surveys show a decrease in the relative abundance of brook trout (Salvelinus fontinalis), as measured by the catch per unit effort, with decreasing pH [Fig. 4.6; Fig. 3.2 in Talbot et al. (1984)].

Smith et al. (1986) report the results of surveys of water chemistry and fish in 234 lakes in Nova Scotia during the summers of 1981-1984. The fish surveys were conducted using experimental gill nets set for approximately 23 hours. The more acidic lakes generally had the lowest species richness of fish. Correlations of fish species richness and water chemistry variables, including pH, aluminum, sulfate, iron, and manganese showed that the water chemistry variables accounted for <40% of the variation in species richness among the lakes. Stepwise multiple regression analysis of fish species vs water chemistry variables "revealed little predictive power" of the variables tested, including pH, aluminum, sulfate, iron, and manganese. The authors concluded that species distribution and species richness among the Nova Scotian lakes surveyed showed no relationship with water chemistry apart from pH. These results are generally consistent with other studies (e.g., Kelso et al. 1986a) that show a decline in the species richness of fish in lakes with declining pH. The influence of pH on fish abundance (i.e., density), however, is less clear.

Frenette et al. (1986) also showed that catch per unit effort for brook trout declined with pH in 37 lakes located in four regions of Quebec. Furthermore, they showed that the relative abundance of brook trout is more highly correlated with calcium concentration than with pH, a result supporting laboratory and field studies which demonstrates that calcium has a mitigating effect on low pH (Brown 1982). The authors indicate that this species had disappeared from three acidic lakes (pH 4.6 to 5.1) with low calcium concentrations in one region (Charlevoix), although they provide no documentation that brook trout were once present in the three lakes. In addition, none of these cited studies on brook trout lakes in Quebec provides evidence of historic changes in pH and losses of brook trout populations resulting from recent acidification associated with acidic deposition. Nevertheless, they do demonstrate the potential impact on brook trout populations of declining pH. The critical pH threshold at which this species is eliminated, however, appears to vary based on evidence from these surveys in Canada.

Brook trout were collected from some lakes in Quebec with a pH <5.0, whereas Beggs and Gunn (1986) found no brook trout in lakes in Ontario with a pH <5.0. This apparent difference in the acid sensitivity of brook trout between Quebec and Ontario may reflect (1) regional differences in the acid sensitivity of this species due to different genetic strains, (2) regional differences in physiological acclimation to acidic conditions, (3) regional differences in aspects of water chemistry other than pH that affect the response of fish to acidic conditions (e.g., differences in calcium or aluminum concentrations or in ligands that complex aluminum), or (4) a combination of these factors. Whatever the cause, this difference indicates spatial variability in the critical pH threshold of a given species.

Synoptic surveys have also been conducted for other taxonomic groups of aquatic organisms. Kelso et al. (1986a) report the results for benthic invertebrate surveys from 198 lakes in Ontario, Quebec, Newfoundland, and Nova Scotia. The results for all regional data combined show that the number of benthic invertebrate species declines with decreasing pH. In contrast, regressions of invertebrate density and biomass vs pH and ANC of lake water were not significant for lakes in most of these regions, implying that the abundance and biomass of aquatic invertebrates is not influenced by acidification. That the number of species is reduced with declining pH whereas the density and biomass show no trend suggests that the loss of some species may be offset by increases in the abundance of at least some of the remaining acid-tolerant species. However, many of the species eliminated at higher pH values are benthic Crustacea (*Mysis*, *Hyalella*, *Pontoporeia*) or molluscs. Replacing these taxa with more acid-tolerant insects or zooplankton is no indication that the food web will function normally.

Zooplankton data from 272 lakes in Ontario, Quebec, and Newfoundland are also reported in Kelso et al. (1986a). The number of zooplankton species in the major groups and for all species combined declines with the decreasing pH of lake water. In general, the number of zooplankton species begins to decline when the pH falls to <6.0, and

the apparent loss of species is more rapid when the pH falls to <5.5 (Kelso et al. 1986a). Regression analyses revealed significant relationships between the number of taxa and pH in lakes in some regions, although the fraction of variation in the number of zooplankton explained by pH alone was relatively low. This suggests that pH alone is not the primary factor regulating the number of zooplankton species in Canadian lakes.

A synoptic survey of aquatic insect communities in eight headwater streams ranging in pH from 4.3 to 6.3 in south-central Ontario was also conducted in 1981 (Mackay and Kersey 1985). The number of genera of aquatic insects generally decreased with decreasing pH. In the most acidic streams (pH 4.3 to 4.5) mayflies were either completely absent or had a reduced species diversity compared with that of streams with higher pHs. These results on the diversity of benthic invertebrate taxa in acidic streams are thus consistent with those for acidic lakes in which the number of invertebrate taxa declined with increasing acidification.

Hall and Ide (in press) sampled the aquatic insect taxa in low alkalinity streams in Algonquin Park, Ontario, that were sampled 48 years ago. At sites where current pH depressions in the spring are low (pH 6.4-6.1), the taxa present in 1984-85 were present during the original survey. In contrast, at sites exhibiting large spring pH depressions (from pH 6.4 to 4.9), several species of mayflies and stoneflies known to be acid-sensitive based on laboratory and field bioassays and field surveys were no longer present. In addition, many of these acid-sensitive species had been replaced by other acid-tolerant mayfly and stonefly species that were not present during the original survey. These shifts in acid-sensitive and acid-tolerant species of aquatic insects suggest that poorly buffered streams in the Algonquin Park region have acidified sometime within the last fifty years.

Evidence from synoptic surveys of surface waters in eastern Canada shows a trend of a decreasing number of species of phytoplankton, zooplankton, benthic macroinvertebrates, and fish with decreasing pH.

The relative abundance of fish (as reflected by the catch per unit effort) also tends to decline with declining pH below pH 6. These empirical relationships implicate acidification as a causal factor responsible for the lower species richness of aquatic communities and lower abundance of fish. This evidence is corroborated by results from laboratory bioassays and in situ bioassays of acid-sensitive and acid-tolerant species of aquatic organisms (Hall et al. 1980; Schindler et al. 1985; Mills and Schindler 1986; Zischke et al. 1983; Ormerod et al. 1987).

4.4 EFFECTS OF EPISODES

To date, more attention has been given to gradual, long-term changes in the acidification of surface waters and to the average annual chemistry of surface waters than to short-term (hourly, daily) changes associated with rainstorms and snowmelt. There is now growing awareness that superimposed on the long-term acidification of surface waters are episodes during which rapid depressions in pH and increases in toxic trace metals can occur (Marmorek et al. 1987). The biological significance of such episodes in Canadian surface waters is assessed in this section.

Depressions in the pH and ANC of lakes and streams in eastern Canada during snowmelt events are well documented (Jeffries et al. 1979, LaZerte and Dillon 1984, Gunn and Keller 1984), and the reader is referred to Sect. 2.6 for a discussion of the extent and probable causes of these changes. Evidence from laboratory bioassays and in situ bioassays indicates that short-term depressions in pH and ANC and changes in other water-quality constituents can stress aquatic organisms and increase mortality (Marmorek et al. 1987).

Evidence that such episodes are adversely affecting aquatic organisms and communities in surface waters is, however, limited both in the United States and in Canada. Harvey and Lee (1982) reported that dead and dying fish, primarily pumpkinseed (*Leposmis gibbosus*), were observed in Plastic Lake, Ontario, during spring runoff from 1979 to 1981. The pH of the inlet streams during spring runoff was as low

as 3.85, but the lake was not as acid (pH of 5.8) during the same period. Although the evidence is circumstantial, Harvey and Lee (1982) suggest that the fish kills were due to acidification.

The lack of documentation of other biological effects during episodes in Canada is probably due in part to a lack of observations of streams and lakes during episodes and to difficulties in observing and documenting stress or mortality of fish (particularly for small, nonmobile life history stages) and other aquatic organisms during peak flows when water in streams and lakes can be turbid. Further, mobile aquatic organisms such as fish may seek refuge in less-acidic areas during these episodes and, hence, avoid toxic conditions. Based on laboratory and in situ bioassays, chemical fluctuations of the magnitude observed can be expected to cause adverse biological effects. There are, however, currently no field programs in Canada specifically designed to extensively monitor fish populations during snowmelt and storm events. Thus, because the acid-base chemistry during episodes attains levels that are acutely toxic to some life stages of some species and because these stages of some species are present in streams, rivers, and lakes in Canada during snowmelt (Gunn 1986), the potential for adverse biological effects exists and is a reason for concern.

4.5 SUMMARY

Historical records of species richness and relative abundance of fish in streams and rivers in eastern Canada show a decline in the number of species and a reduction in the relative abundance of selected species as pH declines below pH 6. Results of laboratory bioassays, in situ bioassays, and field surveys tend to corroborate that acidification may have contributed to the decline in species richness and relative abundance of fish in these systems.

Results of synoptic surveys of lakes and streams in eastern Canada show a trend of decreasing richness of phytoplankton, zooplankton, benthic macroinvertebrate, and fish species as pH decreases. The

relative abundance of fish also declines as pH decreases below 6. These results implicate acidification as a cause of lower species richness and relative abundance. Laboratory bioassays and in situ bioassays tend to support the conclusion that acidification can cause these changes.

Adverse biological effects of episodes (snowmelt, stormflows) on aquatic communities and organisms in eastern Canada are virtually undocumented. There are, however, well-documented depressions in pH and ANC in lakes and streams in eastern Canada during snowmelt. Laboratory bioassays and in situ bioassays of acid-sensitive fish and aquatic invertebrates, nevertheless, clearly indicate the potential for adverse biological effects on aquatic species and communities during such episodes.

5. PREDICTION OF FUTURE EFFECTS OF ATMOSPHERIC DEPOSITION ON SURFACE WATER CHEMISTRY AND BIOTA

(David R. Marmorek)

This section uses modeling results to estimate the future extent of chemical and biological impacts from acidic deposition in eastern Canada. As in previous sections, the focus is on water chemistry, because chemical models have received much more attention than biological models. Section 5.1 describes the mechanistic and empirical models that have been used to predict surface water chemistry, with most of the section devoted to a detailed description of one regional empirical model that forms the basis for projections in Canada. The structure of the model, the uncertainties inherent in its predictions, and the sources of input data to the model are all described in detail.

Section 5.2 presents the results of this regional empirical model for eastern Canada, under current and alternative deposition scenarios. Both uniform percent reductions in deposition and deposition ceilings (or target loadings) are examined. Finally, Sect. 5.3 presents preliminary estimates of the impacts of acid deposition on Ontario fisheries.

5.1 CHEMICAL MODELS

To date, models of the effects of acidic deposition on aquatic chemistry can be classified into two broad groups:

1. empirical models that predict the regional impacts of current or altered levels of acidic deposition on surface water chemistry (Henriksen 1979, 1980; Minns 1981; Thompson 1982; Wright 1983) and

2. detailed mechanistic models that simulate many watershed and lake processes and attempt to mimic the behavior of particular intensively studied watersheds (Christophersen and Wright 1981; Schnoor et al. 1982; Chen et al. 1983; Cosby et al. 1985b).

Both of these types of models have limitations. Empirical models, although yielding predictions of regional-scale impacts on surface

water chemistry, generally contain no physical representation of a watershed, stream, or lake. Detailed mechanistic simulation models typically require far too many input parameters to be useful on a regional scale.

In the last five years, Canadian research scientists have been developing and applying mechanistic models to intensively studied sites and empirical models to regional survey data. The purposes of these investigations are: (1) to understand the hydrochemical processes governing surface water chemistry, and (2) to allow predictions of future status given alternative deposition scenarios.

5.1.1 Site-Specific Mechanistic Models

Mechanistic models of aquatic chemistry that have been recently developed or applied in Canada include the following:

- A hydrochemical model for predicting stream chemistry, similar in structure to the Birkenes/Storgama model of Christophersen et al. (1982, 1984), which was developed at the National Water Research Institute in Burlington, Ontario (Bobba and Lam 1984; Lam and Bobba 1984). It has been applied to three intensively studied watersheds: the Turkey Lakes in the Algoma Region of Ontario (Lam et al. 1986); Harp Lake in the Muskoka-Haliburton Region of Ontario (Bobba et al. 1986); and the Lac Laflamme watershed in northeastern Quebec;
- The Birkenes and Storgama model (Christophersen et al. 1982, 1984), which has been adapted and applied to the Harp Lake watershed (Rustad et al. 1985; Seip et al. 1985);
- A snowmelt chemistry model (Jones, H. G., et al. 1986), developed for and applied to the Lac Laflamme watershed.

To date, these detailed models have been used for two purposes: (1) to determine the minimum level of model complexity required to simulate seasonal and episodic fluctuations in streamflow and stream chemistry, and (2) to make inferences concerning the relative importance of various hydrologic and hydrochemical processes among

watersheds and within years. These modeling activities, however, have not yet attempted long-term, regional predictions nor evaluated alternative deposition scenarios. The only published examination of the effects of altered levels of sulfur deposition in Canada using a mechanistic model is the study by Rustad et al. (1985) at Harp Lake. They found that during snowmelt current pH declines of 0.4 units (pH 5.7 to pH 5.3) would become 0.5 to 0.7 unit declines if sulfur deposition were doubled and would decrease to depressions of 0.1 to 0.2 units if sulfur deposition were reduced by 50%.

The high-quality data collected at the above-described intensively studied watersheds make possible the application of other mechanistic models such as the Model of Acidification of Groundwaters in Catchments (MAGIC) (Cosby et al. 1985a), which are specifically designed to simulate long-term trends.

5.1.2 Regional Empirical Models

Since the MOI (1983), the Canadian Department of Fisheries and Oceans (DFO) has sponsored a series of activities aimed at producing regional estimates of the potential damage to surface water chemistry and inland fisheries that would result from acidic deposition. These activities include modeling studies (Minns et al. 1986; Jones et al. 1984; Jones and Cunningham 1985), lake and stream surveys (Kelso et al. 1986a), and map analyses to estimate the number and area of lakes in eastern Canada (Minns 1984; Kelso et al. 1986a). Unlike the USEPA Direct-Delayed Response Project, which is making regional extrapolations from detailed process models run on intensively studied sites, the Canadian program has been based on simple steady-state models for which regional data already exist.

The only available estimates of the potential regional impacts of acidification on the surface waters and fisheries of eastern Canada are those given in Minns and Kelso (1986). Their estimates are based on a regional model developed by the DFO and described in Jones et al. (1984) and Jones and Cunningham (1985).

5.1.2.1 General Structure

The DFO model consists of both a single-lake site model and a multiple-lake regional model (Fig. 5.1). The site model consists of a set of equations to predict the eventual steady-state chemical status of a single lake (ANC, pH, cations, and sulfate) based on the watershed's morphometry and runoff, current chemistry of the lakewater, observed or assumed levels of sulfate deposition, and assumed values of three other parameters. This site model is embedded in the regional model. The regional model uses frequency distributions for most model parameters and estimates of the number of lakes within each of 36 secondary watersheds east of the Ontario border and south of 52°N. The site model is run many times, once for each "lake type," which consists of a set of parameter values drawn from the frequency distributions, allowing for interparameter correlations (Fig. 5.1). The output of the model includes regional frequency distributions of the estimated original and eventual lake pH (only the latter is shown in Fig. 5.1.).

5.1.2.2 Detailed Site Model Description

The site model is based largely on the empirical models of Henriksen (1979), Thompson (1982), and Wright (1983). Henriksen (1979) presented the concept of whole-lake titration with acid, whereby the ANC of a lake is consumed. Henriksen (1982a) added the concept, called the F-factor, that some fraction of the sulfuric acid falling on the watershed of a lake is exchanged for base cations, with the result that not all the acid deposition in the watershed depletes lake ANC. Wright (1983) presented an extensive application of the chemical balance approach, taking into account the F-factor and using lake sulfate levels as indicators of deposition. These empirical modeling approaches were concerned with the sums of major anion and cation concentrations. Thompson (1982) offered a budget approach in which the output of major ions by watersheds could be computed with allowances made for changes in runoff and acid deposition, but did not include the F-factor.

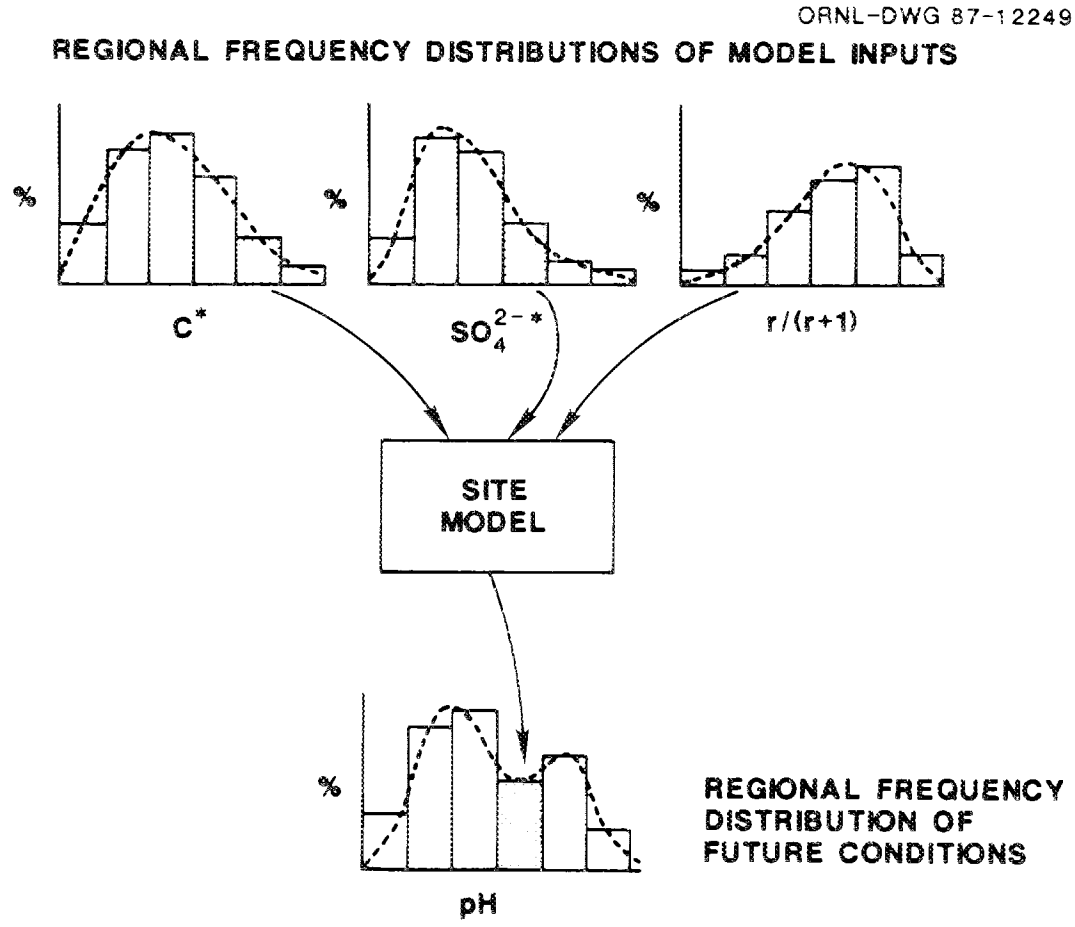

Fig. 5.1. General structure of the overall model. The regional model selects combinations of site characteristics and runs the site model for each combination (shaded bars). By integrating overall observed parameter combinations, a frequency distribution is generated that represents the regional values. The variables are defined in text. Only three of six input frequency distributions are shown. A frequency distribution of model outputs is generated. Source: Marmorek et al. (in prep).

The DFO site model (Jones et al. 1984) generalizes these three approaches to allow for the budgeting of major base cations (Ca and Mg), sulfate, and ANC, with explicit consideration of the F-factor, the role of the watershed-to-lake-area ratio (r = Aw/Ao), and the levels of background lake sulfate (i.e., nonmarine sulfate not attributable to anthropogenic sources). A major difference between the DFO model (Jones et al. 1984) and that of Wright (1983) is DFO's use of sulfate deposition (wet plus dry) as the predictor of lake chemistry, rather than using lake sulfate concentrations as the sole indicator of deposition levels. The site model combines two approaches, back calculation of watershed ANC and back calculation of original ANC, given background sulfate and cation exchange, to predict the original (preacidic deposition) lake ANC.

In the DFO model (Jones et al. 1984), a simple statement of the ANC mass balance, assuming no sulfate retention, is combined with the premise that there is a proportionality between ANC and major ions that is altered by sulfate loadings and cation exchange. The resulting equations can be rearranged to allow the prediction of steady-state ANC, ANC_∞, for a steady-state level of sulfate deposition, SD_∞:

$$ANC_\infty = \alpha\, C_t^* - \left[\alpha\left(\frac{r}{r+1}\right) F_W \quad (SO_4^* - \beta)\right] - \left[1 - \alpha\left(\frac{r}{r+1}\right) F_W\right] \frac{SD_\infty}{P \bullet WR} \quad , \qquad (1)$$

where

- ANC_∞ = steady-state ANC (eq L^{-1}),
- ANC_0 = original ANC, preacidic deposition (eq L^{-1}),
- r = Aw/Ao,
- Aw = watershed area excluding the lake area (m^2),
- Ao = lake area (m^2),
- P = annual precipitation (L m^{-2} yr^{-1}),
- WR = fraction of precipitation remaining after evapotranspiration,
- α = the proportionality between ANC and C_t (after Wright 1983),
- C_t^* = current concentration of nonmarine (Ca + Mg), (eq L^{-1}),

F_W = empirical factor accounting for cation exchange with hydrogen ion in the terrestrial watershed (after Henriksen 1982b and Wright 1983),

SO_4^*, β = current and original (background) concentrations of nonmarine sulfate (eq L^{-1}), respectively, and

SD_∞ = sulfate deposition at steady-state [negative ANC (eq m^{-2} yr^{-1})].

The first term in the equation represents original ANC, the second term corrects for cation exchange, and the third term reflects the titration of the original ANC caused by changes in sulfate deposition. Because the level of background sulfate (β) strongly determines the proportionality (α) of ANC and cations (Wright 1983), the most important factors in determining a lake's response to increased sulfate deposition are β and F_W.

Estimates of β can be made by examining the mean SO_4^{2-} concentrations in reference area lakes that receive only low levels of deposition (Wright 1983). Within eastern Canada, the only such areas are northwestern Ontario and Labrador, both of which annually receive approximately 10 kg ha^{-1} of wet SO_4 deposition. The mean SO_4^{2-} for Labrador lakes in the National Inventory Survey is 25.4 $\pm$ 1.5 (SE; n = 75). For northwestern Ontario, however, the mean SO_4^{2-} is 101.6 $\pm$ 5.4 (SE; n = 44). In Jones et al. (1984), simulations to predict ANC_∞ used two extreme estimates for β: 20 and 100 μeq L^{-1}. The same value for β was used for all lake types during each run, with the added constraint that whenever ($SO_4^*-\beta$) was negative, it was reset to zero.

Empirical estimates of F_W require long-term series of high-quality water chemistry data from lakes that have experienced a change in levels of acidic deposition. Unfortunately, most potential data sets have either unreliable historical measurements or are of insufficient duration to show significant changes in deposition. Henriksen (1982b) used two indirect approaches to estimate F_W:

1. comparisons of historical (preacidification) and recent (postacidification) data from both Scandinavian and North American systems, and

2. regressions of ($Ca^* + Mg^*$) against (SO_4^{2-*}) for lakes in southern Norway and the Sudbury area of Ontario (i.e., substituting spatial trends for temporal ones and assuming that (SO_4^{2-*}) was proportional to deposition).

Henriksen concluded that 0.4 was probably the maximum value for F_w. This conclusion depends on various critical assumptions, such as the original ($Ca^* + Mg^*$) and (SO_4^{2-*}) concentrations, the effects of changing hydrology on chemical concentrations, and the quality of older data. Henriksen's analyses suggest that his conclusion is probably valid for the mean value of F_w (i.e., if a constant value is applied for all lakes), but that individual lakes may show values of F_w higher than 0.4. Wright (1983) made further indirect estimates and surmised that a range of 0 to 0.4 for F_w was "probably reasonable"; his analyses, however, did not provide any definite proof for this statement. For a lake that was experimentally acidified with direct additions of sulfuric acid (Lake 223, Experimental Lakes Area, Ontario), the ratio of increases in base cations to increases in sulfate during the first eight years of manipulation was 0.4 (Cook et al. 1986). This value is probably an underestimate for lakes experiencing atmospheric deposition, because it does not include changes in weathering in the watershed.

Finally, Dillon et al. (1986) estimated a value of F of 0.64 for Swan Lake, a lake in the Sudbury Area that experienced a substantial decrease in both sulfate and base cation concentrations (62% and 45%, respectively). Though this system was recovering rather than acidifying, the model of Jones et al. (1984) and other similar models assume that the acidification process is reversible, so the estimate from Swan Lake is relevant. However, base cation concentrations in Swan Lake (514 $\mu eq\ L^{-1}$ in 1977; Dillon et al. 1986) are considerably higher than in most regions with low-ANC systems. Wright et al. (1986) used simulation runs of the MAGIC model to demonstrate theoretical support for the hypothesis that F_w is positively correlated with

base cation concentrations. This suggests that F_W of 0.64 for Swan Lake is at the upper end of the range of F_W for lakes vulnerable to acidification; the mean value for regions with such lakes is probably much lower.

In simulation runs for eastern Canada, Jones et al. (1984) assumed two mean values for F_W (0 and 0.5) and applied each of these to all lake types in successive model runs. Therefore, together with two extreme values for β, four different parameter combinations were simulated. Table 5.1 lists the information sources for all model parameters. The effect on model parameters of underestimating or overestimating key parameters (i.e., α, β, or F_W) is not intuitively obvious. Rather, it depends on whether the lake is acidifying $[(SD_\infty/P \bullet WR) > (SO_4^{2-} - \beta)]$ or recovering $[(SD_\infty/P \bullet WR) < (SO_4^{2-} - \beta)]$ (Jones et al. 1984).

More recent modeling work in Canada (Jones, M. L., et al. 1987) has assumed that F_W is linearly proportional to estimated original ANC (ANC_0), which may be a more reasonable assumption because some lakes are then assigned F_W greater than 0.5. Dynamic models that vary F_W over time have been neither developed nor applied on a regional scale in Canada.

Figure 5.2 shows the order of calculations in the site model. Once ANC_∞ has been computed using Eq. (1), the steady-state pH (pH_∞) is calculated based on (1) a set of curves describing ANC-pH relationships for surface waters of different DOC levels (described in Minns et al. 1986 and based on Driscoll 1980), and (2) an empirically derived function of pH and DOC (Jones et al. 1984) to correct the above function for underestimates of pH at low ANCs (Jones et al. 1984).

Model predictions are generally stated in terms of the number of lakes with a predicted steady-state pH of less than 5, since few fish species inhabit lakes with a pH <5. More recent modeling analyses provide output as a cumulative frequency distribution of pH values (Jones et al. 1987). For fish species that have been sufficiently studied to generate curves of the probability of species loss vs mean pH, such as brook trout in the Adirondacks (Baker and Harvey 1984;

Table 5.1. Sources of estimates for model parameters, assumed values, and potential uncertainty

Parameter	Source/assumed values	Uncertainty
SO_4^*, C_t^*, DOC	Frequency distributions for secondary watersheds (data as in Jeffries 1986)	Small sample sizes in some secondary watersheds mean large uncertainty in frequency distributions; however, most of these watersheds have low deposition
α, β	Water chemistry surveys from NW Ontario (α = 0.67; $\bar{\beta}$ = 100 µeq L^{-1}) used for Ontario 2° watersheds; Surveys from Labrador (α = 0.91; $\bar{\beta}$ = 20 µeq L^{-1}) used for other provinces	Assumes that areas with moderate-to-high acidic deposition originally had $[SO_4^*]$ similar to remote areas. Current $[SO_4^{2-*}]$ in Ontario is significantly higher than in other provinces (see Table 2.3).
SD_{∞}	For "current deposition" scenarios, 1980 wet sulfate deposition is multiplied by 1.25 to account for dry deposition. For other scenarios, assumed value is used for wet deposition; 1.25 multiplier is still used to account for dry deposition	Dry deposition underestimated; recently measured values range from 1.67 in SW Ontario to 1.18-1.25 in remote areas (Barrie and Sirois 1986)
F_w	Assumed to vary from 0 to 0.5 (Henriksen 1982b; Wright 1983); both values applied to all lakes	Maximum F_w probably >0.5 for single lakes. Dillon et al. (1986) estimated F_w = 0.67 for Swan Lake in Sudbury area. Regional mean F_w probably <0.5 in sensitive regions. Wright et al. (1986) suggest F_w correlated with original cation concentrations
P, WR	Hydrological Atlas of Canada (1978); long-term averages	Long-term average value of P*WR may not be valid for the particular year's data used for SD_{∞}. More accurate estimate requires measurements from same year
r (Aw/Ao)	Minns (1984); DFO Counts and Measures data (unpublished)	All lakes assumed to be headwater lakes. Further analyses indicate predictions are not sensitive to this assumption (Jones and Wedeles 1986)

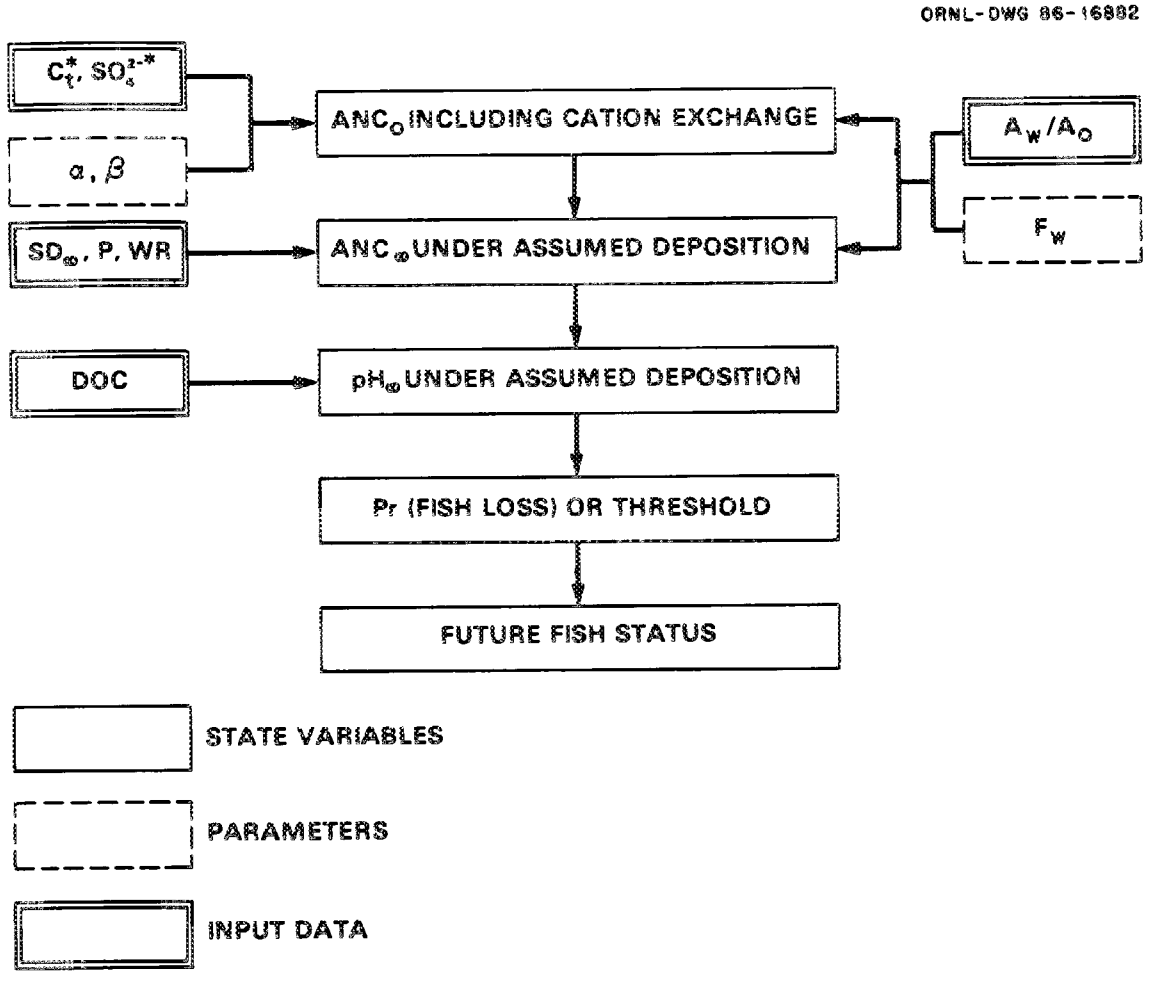

Fig. 5.2. Flowchart showing how the input data to the model and assumed parameter values are combined to yield the model's outputs. Both the method and the parameters used to convert ANC_∞ to pH_∞ are described in Minns et al. (1986). Six other parameters (all equilibrium constants) are used.

Reckhow et al. 1985) and lake trout in Ontario (Beggs et al. 1985), it is possible to estimate the probable number of lakes that will lose fish populations as a result of decreased pH. Some approaches to this problem are presented in Sect. 5.3.

Table 5.2 summarizes some of the simplifying assumptions made by the site model and their consequences for model predictions. Though sulfate retention and reduction are not included in the model, this should not greatly affect the accuracy of steady-state predictions of lake chemistry. Once soils reach equilibrium with acidic deposition, surface waters will also. Sulfate retention occurring at a given site could be responsible for the predicted steady-state pH being lower than that currently observed.

5.2 CHEMICAL MODEL APPLICATION TO AQUATIC SYSTEMS IN CANADA

The results of model application are presented in Table 5.3 and in Fig. 5.3. Table 5.3 includes predictions of the number and percentage of lakes projected to have an eventual steady-state pH of <5 for two assumed values of F_w (0 and 0.5), one set of values for α and β for each region, and six acidic deposition scenarios: (1) 1980 levels of deposition, (2) a 40% reduction in deposition in all subregions, (3) a 20% increase in deposition in all subregions, (4) a target loading of 13.5 kg ha^{-1} SO_4^{2-} (wet and dry), (5) a target loading of 18 kg ha^{-1} SO_4^{2-} (wet and dry), and (6) a target loading of 27 kg ha^{-1} SO_4^{2-} (wet and dry). Proportional reductions in deposition were implemented by multiplying the estimated total current deposition by the same fraction for each subregion. Target loadings were implemented by changing the level of deposition to the target loadings in all subregions where deposition currently exceeds the target; in subregions where current deposition is less than the target, 1980 levels were maintained. Figure 5.3 represents the target loading simulations as a separate "threshold deposition control axis."

Also included in Table 5.3 are the number and percentage of lakes currently observed to have a pH of <5. Figure 5.3 presents the highest and lowest estimates of the number of lakes with a predicted

Table 5.2. Major assumptions in the Department of Fisheries and Oceans (DFO) model and their consequences[a]

Model assumptions that might cause an underestimate of the extent or magnitude of damage to surface waters and fisheries	Model assumptions that might cause an overestimate of the extent or magnitude of damage to surface waters and fisheries
• The original acid neutralizing capacity (ANC) of watersheds (eq ANC m^{-2}) is not reduced by acidic deposition	• ANC generation within the lake is not included, although the DFO model has been revised (Jones, et al. 1987) to include the model of ANC generation of Kelly et al. (1987)
• Episodic pH depressions, which may have serious consequences for fisheries, are not simulated	• All sulfate in wet deposition is assumed to be associated with H^+. The revised model of Jones et al. (1987) deducts an amount of sulfate equal to base cations from total sulfate deposition
• Model output of the number of lakes with pH <5 does not reflect acidification to pH 5-6, where aquatic effects do occur. [More recent analyses (Jones et al. 1987) indicate that the number of lakes acidified to pH <5.5 is 2-3 times the number acidified below pH 5.0]	
o underestimates of SD_{∞} (e.g., underestimating dry deposition)	o overestimates of SD_{∞}
o overestimates of runoff (P WR)	o underestimates of runoff (P WR)

[a]Effects of parameter uncertainties (open circles) are shown only for those parameters that have a consistent effect on model predictions; other parameters (α, β, F_W) have interactive effects.

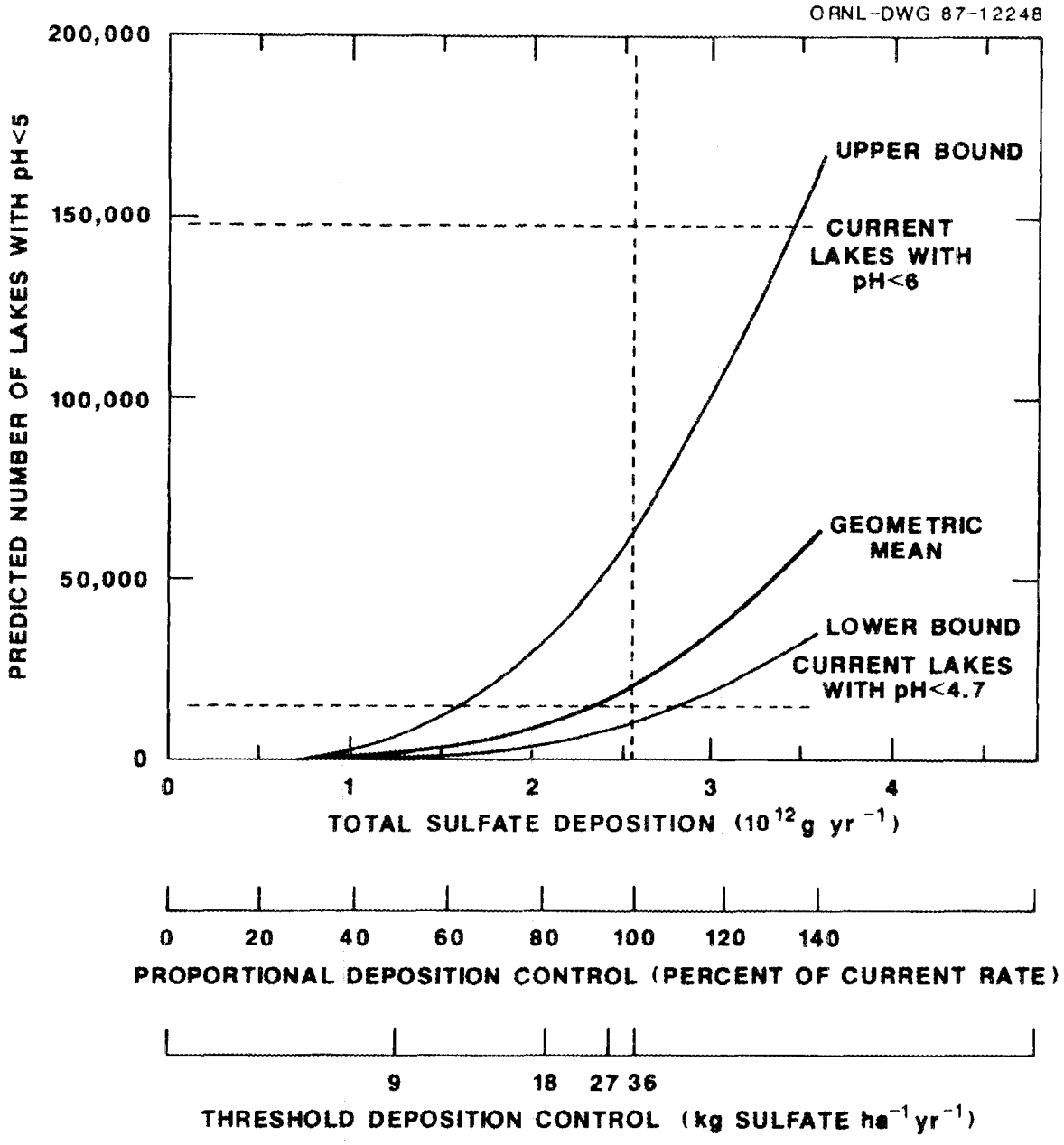

Fig. 5.3. Predicted relationship between the number of lakes with pH <5 and total sulfate deposition in eastern Canada, south of 52°N. The lower and upper bounds represent the minimum and maximum damage predictions from four model runs, using a wide range of model parameters. The vertical line of dashes indicates the level of deposition that is currently observed. The proportional deposition control axis assumes proportional reductions in deposition in all secondary watersheds (see text). The threshold deposition control (i.e., target loading) axis lowers deposition to the threshold or to the current deposition, whichever is lower, in all secondary watersheds (see text). Source: Minns and Kelso (1986).

steady-state pH of <5 for four parameter combinations [two alternative values for β (20 and 100 μeq L^{-1}) for each of the two F_w assumptions]. The geometric mean of the four predictions is also shown. Because two extra parameter combinations were used in Fig. 5.3, its range of predictions is larger than those in Table 5.3. Model predictions differ from currently observed conditions whenever a lake's sulfate concentrations are not in equilibrium with the level of sulfate deposition (i.e., $[(SD_{\infty}/P\ WR) = (SO_4^{2-*} - \beta)]$).

Though the uncertainty in parameter values leads to considerable uncertainty in the predicted damage, the model's predictions are valuable for placing preliminary bounds on the potential extent of acidification. Under current deposition rates, the predicted number of lakes that will eventually have steady-state pH values of <5 ranges from 10,000 to 60,000 lakes (Fig. 5.3), using four extreme sets of parameter values, or 10,000 to 36,000 using the two most reasonable sets of parameter values (Table 5.3), as compared with the 12,000 lake estimate based on extensive surveys of present conditions (Table 5.3; see also Sect. 2.4). The predictions of the number of lakes decreasing to pH values below 5 under current deposition are within the range of survey-based estimates of current conditions in Quebec and the Maritimes but exceed the survey estimates in Ontario (Table 5.3). More recent modeling analyses (Jones et al. 1987) indicate that the predicted number of Ontario and Maritime lakes that will attain a pH <5.5 under the current deposition is roughly twice the predicted number of lakes that will reach pH <5.0. In Quebec, the predicted number of lakes at a pH <5.5 is about three times the predicted number of lakes with pH <5. Thus, estimates of the number of lakes experiencing a pH decline to below 5.5 are two to three times higher than those declining to pH <5.0. As discussed in Sect. 4, experimental acidification studies have shown that considerable damage to aquatic biota can occur when pH decreases below 5.5 to 5.7 (Schindler et al. 1985).

The province with the largest number of lakes experiencing pH change is Ontario (5,600 to 15,400 lakes)(Table 5.3). The subregions of Ontario with the greatest predicted change are in the south-central part of the province: the Muskoka-Haliburton area east of Georgian Bay;

Table 5.3. Observed (estimated from surveys) and predicted number (%) of lakes with pH <5, by region, for two assumed values of F_w and six acid deposition scenarios. Target loadings, expressed as kg SO_4^{2-} ha^{-1} $year^{-1}$, include both wet and dry deposition

Region	Total number of lakes	Number (%) of lakes with pH <5 estimated from surveys[a]	Assumed parameter values			Number (%) of lakes predicted to have eventual steady-state pH <5 under stated deposition scenario					
							Change in deposition		Target loading (kg SO_4^{2-} ha^{-1} $year^{-1}$)		
			α	β	F	Current deposition	40% reduction	20% increase	13.5	18	27
Ontario	137,415	1,649 (1.2)	0.67	100	0	15,417 (11)	4,843 (3.5)	21,732 (16)	1,145 (0.8)	3,511 (2.6)	12,988 (9.5)
			0.67	100	0.5	5,629 (4.1)	224 (0.2)	9,407 (6.9)	212 (0.2)	616 (0.4)	2,751 (2.0)
Quebec	229,458	3,691 (1.6)	0.91	20	0	11,103 (4.8)	801 (0.35)	25,005 (11)	1,994 (0.9)	5,758 (2.5)	10,019 (4.4)
			0.91	20	0.5	915 (0.4)	0 (0)	4,797 (2.1)	773 (0.3)	773 (0.3)	871 (0.4)
Maritimes	243,245	7,165 (3.0)	0.91	20	0	9,464 (3.9)	1,726 (0.7)	15,350 (6.3)	2,753 (1.1)	6,737 (2.8)	9,464 (3.9)
			0.91	20	0.5	3,463 (1.4)	1,123 (0.5)	5,876 (2.4)	1,351 (0.6)	2,774 (1.1)	3,463 (1.4)
TOTAL	610,108	12,505 (2.1)			0	35,984 (5.9)	7,370 (1.2)	62,087 (10)	5,892 (1.0)	16,006 (2.6)	32,501 (5.3)
					0.5	10,007 (1.6)	1,347 (0.2)	20,080 (3.3)	2,336 (0.4)	4,163 (0.7)	7,085 (1.2)

[a]Based on survey data summarized in Jeffries (1986) and scaled up using Counts and Measures data (Department of Fisheries and Oceans, Canada).

Sources: Jones et al. 1984, Jones and Cunningham 1985.

Algonquin Park; and that area between Lake Ontario and the Ottawa River that is underlain by the Canadian Shield. The range in the number of lakes predicted to decrease in pH in Quebec, 900 to 11,100 lakes (Table 5.3), is large because the lake water chemistry in these areas is sensitive to changes in F_w. Most of the lakes predicted to experience pH declines are located in the area west of Quebec City and in the area north of the entrance to the St. Lawrence River. Significant portions of the total number of lakes in both Nova Scotia and New Brunswick are included in the estimates for the number of lakes that will reach a pH <5.0 in the Maritimes (Table 5.3), but by far the largest number of lakes that will have pH declines are in northern Newfoundland. Because this subregion contains 128,000 lakes, even if a relatively small proportion of these acidify, the estimate is increased substantially.

Predictions from the model indicate that substantial decreases in the extent of acidification are possible with reduced deposition. Conversely, higher rates of acidic deposition could disproportionately increase the number of acidified systems. Table 5.3 indicates that a 40% reduction in deposition decreases the predicted number of lakes in eastern Canada with a pH of <5 by 80% to 86% (9,000 to 29,000 lakes). On the other hand, a 20% increase in acid deposition is predicted to cause 72% to 100% more lakes to acidify (10,000 to 26,000 lakes). The reason for these dramatic nonlinearities is that a very large proportion (about half) of the lakes in eastern Canada have ANCs less than 50 $\mu eq\ L^{-1}$ (see Sect. 2 of this report) and are therefore close to an acidification (or recovery) threshold.

Substantial nonlinear reductions in predicted damage are also evident as the target loading (or threshold control) imposed on the model is lowered. This is clear from the nonlinear scale of "threshold deposition control" in Fig. 5.3, and the three rightmost columns of Table 5.3. Changing the target loading by 33%, from 27 to 18 kg $SO_4^{2-}\ ha^{-1}\ yr^{-1}$, decreases the predicted number of acid lakes by 41% to 51% (3,000-16,000 lakes). A further 25% reduction in the target loading, from 18 to 13.5 kg $SO_4^{2-}\ ha^{-1}\ yr^{-1}$ (Table 5.3),

lowers the predicted number of acidic lakes by 65% to 67% (400 to 2,400 lakes). The worst-case assumptions illustrated in Fig. 5.2 yield substantially greater benefits from deposition reductions. Note that the model predicts no lakes with pH <5 under a zero deposition scenario. This prediction may be an underestimate for regions with many highly colored lakes having low base cation concentrations. In other regions, however, the prediction may be reasonable. For example, the NIS survey (Kelso et al. 1986a) found no lakes with pH <5 in a sample of 77 low-ionic-strength headwater lakes in a secondary watershed in Labrador that receive less than 10 kg SO_4^{2-} ha^{-1} $year^{-1}$.

It should be stressed that the model includes both wet and dry deposition in the input loading level, so that target loadings expressed as wet deposition alone would be lower by between 20% to 50% (see Table 5.1). Isopleths of wet sulfate deposition in 1980 are shown in Fig. 3.2.

The various deposition control scenarios indicate that neither a 30% reduction in deposition everywhere nor a limitation of 20 kg SO_4^{2-} ha^{-1} $year^{-1}$ (current control objective) would reduce the worst-case results below the existing level of acidification. A 40% reduction everywhere or a 12 kg SO_4^{2-} ha^{-1} $year^{-1}$ limitation would achieve that result. Loucks and Glass (1984) indicated that a limit of 12 kg SO_4^{2-} ha^{-1} $year^{-1}$ was necessary to protect the most sensitive freshwaters. Target loadings recommended by other investigators are listed in Table 5.4. Nonetheless, the current control objective (20 kg SO_4^{2-} ha^{-1} $year^{-1}$) would bring about some improvement.

5.3 FISHERIES MODELS AND THEIR PREDICTIONS

Until recently, regional estimates of fisheries impacts were not available for eastern Canada. Minns (in press) has recently developed preliminary estimates of the effect of changes in lake acidity in Ontario on three indices of the health of fisheries:

Table 5.4. Summary table of recommended precipitation pH levels or sulfate target loadings to prevent lake acidification

Source	Sulfate target loading or pH standard	Comments
Almer et al. 1978	15-17 kg ha^{-1} $year^{-1}$ (total)	Lakes with acid-neutralizing capacities (ANCs) between 100-200 µeq L^{-1} would lose ANC and decline in pH. Very sensitive lakes would be expected to acidify
	9-12 kg ha^{-1} $year^{-1}$ (total)	Very sensitive lakes would be protected from acidifying; no degradation (loss of ANC or pH) in lakes with ANCs between 100-200 µeq L^{-1}
Henriksen 1980	20 kg ha^{-1} $year^{-1}$ (wet)	Majority of sensitive lakes would not acidify; this loading is associated with a precipitation pH of 4.5. Highly sensitive lakes would most likely acidify
	10-12 kg ha^{-1} $year^{-1}$ (wet)	Highly sensitive lakes with ANCs less than 50 µeq L^{-1} would be protected from acidification
Brydges and Neary 1984	20 kg ha^{-1} $year^{-1}$ (wet)	The majority of sensitive lakes will not acidify. Lakes with ANCs less than 50 µeq L^{-1} may acidify
	<15 kg ha^{-1} $year^{-1}$ (wet)	Sensitive lakes would be protected from acidifying. The majority of sensitive lakes would not show any significant effects from acid deposition
Wright 1983	pH 4.7	Protects lakes with Ca + Mg concentrations greater than 40 µeq L^{-1} from acidifying
Oppenheimer 1984	18 kg ha^{-1} $year^{-1}$ (wet)	The majority of the sensitive lakes will be protected from acidifying. Very sensitive lakes may acidify
	<18 kg ha^{-1} $year^{-1}$ (wet)	Very sensitive lakes will be protected from acidifying. The majority of sensitive lakes will probably not show any major effects from acid deposition
Rogalla and Brezonik 1986	pH 4.58-4.78	Lakes with ANCs greater than 45 µeq L^{-1} protected from acidifying
Schnoor et al. 1986	pH 4.6	The most sensitive lakes (seepage lakes) protected from acidifying
MPCA 1985	pH 4.3-4.4 (14-15 kg ha^{-1} $year^{-1}$, wet)	Protects sensitive lakes with ANCs greater than 60 µeq L^{-1} from acidifying. Lakes with ANCs less than 60 µeq L^{-1} will most likely acidify
	pH 4.7 (11 kg ha^{-1} $year^{-1}$, wet)	Protects the very sensitive lakes (ANCs between 40-60 µeq L^{-1}). Prevents loss of ANC and pH in lakes with ANCs between 60-100 µeq L^{-1}

Source: Minnesota Pollution Control Agency (MPCA) 1985.

1. species richness;
2. presence-absence; and
3. production.

Each of these estimates will be discussed in turn.

The species richness predictions are based on a multiple regression equation developed from the Lake Inventory Database of the Ontario Ministry of Natural Resources (Minns 1981). The equation predicts the number of species in a lake as a function of these variables: lake area, species richness in tertiary watersheds (to reflect decreases in richness with latitude), elevation, mean depth, survey year (to reflect the steadily improving quality of fish sampling), and pH. These variables explained 48.4% of the observed variation among lakes in fish species richness.

To apply the richness model, Minns (in press) divided the observed range of lake areas in Ontario into 20 area intervals and then used the data from the Ontario Lake Inventory for all independent variables except pH and survey year. The pH was predicted from the chemistry model of Jones et al. (1987) (a more recent version of the model described in Section 5.2). The survey year was assumed to be 1980. The model predicts an average of 4.35 species per lake before the Industrial Revolution in North America, 4.16 species per lake at steady-state under 1980 levels of deposition (a 4.3% decline), and 4.12 species if deposition were to increase by 10% (an additional 0.8% resource loss). Canadian emission control programs (decreasing emissions by 38%) would, according to this model, produce recoveries of 35.5% of the total number of fish species "lost" as a result of acidic deposition.

Minns' presence-absence model estimates the number of lakes with a given species as:

$$N_j = N_T * \sum_{i=1}^{n} p_{1,i} * p_{2,i}$$

where i = pH interval index (0.5 pH units);
n = number of pH intervals
N_j = number of lakes with species j;
N_T = total number of lakes;
$P_{1,i}$ = proportion of lakes in pH interval i (determined from chemical model predictions); and
$P_{2,i}$ = probability of encountering species j in a lake with its pH in interval i (derived from the Ontario Lake Inventory).

This model is similar to the model developed by Reckhow et al. (1985), though the use of discrete intervals rather than a continuous function probably makes Minns' approach less precise.

The results indicate that for species with limited distribution in waters of pH <6.4, 1980 deposition rates would eventually cause the loss of these species from 5.7% to 8.6% of Ontario lakes. The range of percentages reflects differing responses and distributions of different species (lake trout, lake whitefish, northern pike, common shiner, blacknose shiner, and walleye). A 10% increase in deposition would worsen this damage to 7.2% to 9.7% of the lakes, whereas Canadian emission controls would reduce the damage to 3.4% to 6.5% for all species but common shiners, allowing total recovery of this species.

For species well distributed in waters of pH <6.4 (brook trout, white sucker, northern redbelly dace, pumpkinseed, yellow perch, and fathead minnow), estimates of species lost were more variable (1.9% to 6.8%). Emission controls resulted in reductions in damage similar to those observed for the group of less acid-tolerant species, though for fathead minnows the model predicts increased losses with emission controls. This result may be an artifact caused by discontinuous encounter probability distributions, or it could reflect implicit species substitutions.

The third model developed by Minns (in press) was based on fish production data from 100 to 200 lakes in the Turkey Lakes watershed. He found that fish production (in kg ha^{-1} $year^{-1}$) could be empirically predicted from lake ANC (in $\mu eq\ L^{-1}$):

$$\text{production} = 7.126 + 0.07624 * \text{ANC} \qquad (r^2 = 0.31).$$

Applying this equation to all Ontario lakes yielded the following results:

1. the 1980 level of deposition would cause a 2.3% decrease in production;
2. increased deposition (10% higher) would raise this estimate of species lost to 2.5%; and
3. Canadian emission controls would restore 20% and 54%, respectively, of the lost production.

Although these regional fisheries models contain considerable uncertainties due both to limited data sets and to low predictive power of empirical relationships, they do offer a rough estimate of the magnitude of damage to Ontario fisheries. Unfortunately, the data base outside Ontario is not as extensive. Further work is necessary to improve both the data bases outside Ontario and to quantify the uncertainty inherent in the Ontario damage estimates.

5.4 SUMMARY

Regional empirical models have been used to estimate the expected changes in surface water chemistry in the lakes of eastern Canada that would result from current (1980) levels of acidic deposition and also that would result from alternative deposition scenarios. These models predict the steady-state condition of lakes, based on estimates of the number and area of lakes within each subregion and their current chemical condition. Estimates of the rate of change in surface water chemistry on a regional scale are not possible with these steady-state models.

Model results show that substantial damage could occur under current (1980) levels of deposition. Once the lakes reach steady-state with the 1980 levels of deposition, 10,000 to 36,000 lakes are predicted to have a pH <5, using the two sets of parameters combinations, or 10,000 to 60,000 lakes using four sets of parameters combinations. More than twice this number of lakes are predicted to reach a pH <5.5. Increases or reductions in deposition cause disproportionately large changes in the predicted number of acidic

lakes. This model behavior arises because a large proportion (about half) of the lakes in eastern Canada have an ANC of less than 50 $\mu eq\ L^{-1}$ and, because of the nature of the relationship between ANC and pH small changes in the level of deposition can make a substantial difference in the number of lakes with predicted pH <5. Similarly, nonlinear reductions in predicted damage are evident as a simulated target loading is lowered. Overall, the various scenarios indicate that neither a 30% reduction in deposition everywhere nor a limitation of 20 kg SO_4^{2-} ha^{-1} $year^{-1}$ (current control objectives in Canada) would reduce the worst-case results below the existing level of acidification, although some improvement would occur. A 40% reduction or a 12 kg SO_4^{2-} ha^{-1} $year^{-1}$ limitation would, however, reduce the worst-case results. Estimates of damage to fisheries are preliminary and available only for Ontario. These models suggest that at steady state, 1980 levels of acidic deposition have decreased the species richness, probability of presence, and production of Ontario fisheries by 4.3%, 5.7 to 8.9%, and 2.3%, respectively. Emission controls in Canada would bring about the recovery of about 35% of the lost species richness, roughly a third of the "lost" probability of presence, and 20% of the lost production.

6. RECOVERY OF AQUATIC SYSTEMS FROM ACIDIFICATION
(David R. Marmorek, Robert B. Cook, and Robert S. Turner)

6.1 CONCEPTUAL FRAMEWORK

Aquatic systems will recover from the effects of acidic deposition after emissions of SO_2 and NO_x have been reduced; in question are the rate and extent of recovery and whether recovery will result in the same biological community or functional characteristics that existed prior to acidification. Recovery may be the result of either of two conditions: (1) the reduction of acidic inputs to the system, or (2) the introduction of substances to the system that neutralize acidity or increase the rate of ANC production. In the first condition, the major issues are the rate and the extent of both chemical and biological recovery. In the second condition, the major issues are the rate and the extent of biological recovery following the rapid return of pH to circumneutral values and the continuing costs of maintaining circumneutral pH.

Regardless of whether the chemical recovery was the result of changes in deposition or liming, the criteria used to judge whether a system has recovered are a key issue. Cairns et al. (1977) offer two useful definitions of restored ecosystems:

1. restoration to original functional and structural conditions [although the elements (species) that comprise the structure may not be the same as those present originally]; and
2. restoration to the original structure and function (with the same species present).

Most of the literature on chemical and biological recovery describes the observed changes and potential mitigative measures. There is, however, a need to make an explicit statement as to which of these two conditions represents the goal of environmental managers responsible for the restoration of acidified systems.

If acidic inputs to an acidified system are reduced, the natural processes of ANC generation, such as soil and rock weathering and in-lake bacterial sulfate reduction, will eventually increase the ANC and pH of the system. The rate and extent of this increase depend on many factors, including the magnitude of the deposition reduction and numerous lake-watershed characteristics.

If a mitigation program such as lake or watershed liming is undertaken, the system will increase in ANC at the rate at which the ANC-generating substance (such as lime) is applied or the rate at which it reacts with the water. Mitigative measures will result in a long-term increase in ANC only if they are applied continually and at a rate exceeding the rate of acidic deposition.

Section 6.2 discusses the evidence that exists for chemical and biological recovery in areas where either deposition reduction has occurred or lakes have been experimentally manipulated.

6.2 EVIDENCE FOR CHEMICAL AND BIOLOGICAL RECOVERY

6.2.1 Evidence for Chemical Recovery from Deposition Reductions

Investigations of the chemical effects of reductions in deposition are difficult because there are few long-term records of surface water chemistry. Nevertheless, studies conducted in areas where deposition has decreased are invaluable, because they provide the strongest empirical evidence either for or against current acidification models and hypotheses. Of particular interest are changes in surface water sulfate concentrations, pH, cation export rates, and organic acid concentrations. Cation export rates are expected to decrease as deposition declines. The F-factor [the ratio of the change in (Ca + Mg) to the change in lake SO_4^{2-}], is positive or zero and has an important effect on the predictions of most steady-state models (Henriksen 1982a; Wright 1983; Jones et al. 1984). These models hypothesize that as deposition declines, lake pH will recover. Functionally, F acts in those models to constrain the change in lake

ANC for a given change in deposition. The lower the value of the F, the greater is the predicted increase in lake ANC for a given reduction in deposition. The rate of lake chemistry recovery will also depend on the capacity of the soils to supply base cations and on whether this capacity has changed as a result of atmospheric deposition.

An alternative hypothesis is that reductions in acidic deposition will have no effect on lake pH. According to Krug and Frink (1983), a reduction in strong acid deposition will induce the release of soil humic acids in nearly equivalent amounts with "little or no measurable change in pH."

The two areas of Canada for which long-term records exist and where deposition reductions have occurred are Nova Scotia and the Sudbury area of Ontario. In Nova Scotia, a decrease in sulfate deposition over a 5- to 10-year period has coincided with decreased concentrations of sulfate in rivers and increased ANC and pH (Thompson 1986). Runoff-corrected cation export rates have remained constant. Figure 6.1 shows the decrease in sea-salt-corrected sulfate yields of eleven Nova Scotia rivers over the period 1971-73 to 1981-82. The two sets of sulfate measurements, performed by methods that yield comparable results (Thorin and ion chromatography; Thompson 1986), indicate decreases in sulfate yield ranging from 40% to 60%. The pH increases associated with these declines in sulfate yields are illustrated in Fig. 6.2. As would be expected from the shape of the bicarbonate titration curve (Stumm and Morgan 1981), the largest pH increases occurred at the pH range 4.9 to 5.8. The magnitude of pH increases was limited by pH buffering by organic anions and aluminum (Thompson 1986).

In the Sudbury area, both large-scale surveys (Keller and Pitblado 1986) and site-specific, intensive studies (LaZerte and Dillon 1984; Dillon et al. 1986; Hutchinson and Havas 1986) provide a detailed picture of surface water responses to reductions in deposition. Emissions of SO_2 from Sudbury smelters have substantially declined since the 1960s: 4240-7034 tonne/d during the period 1960 to 1969,

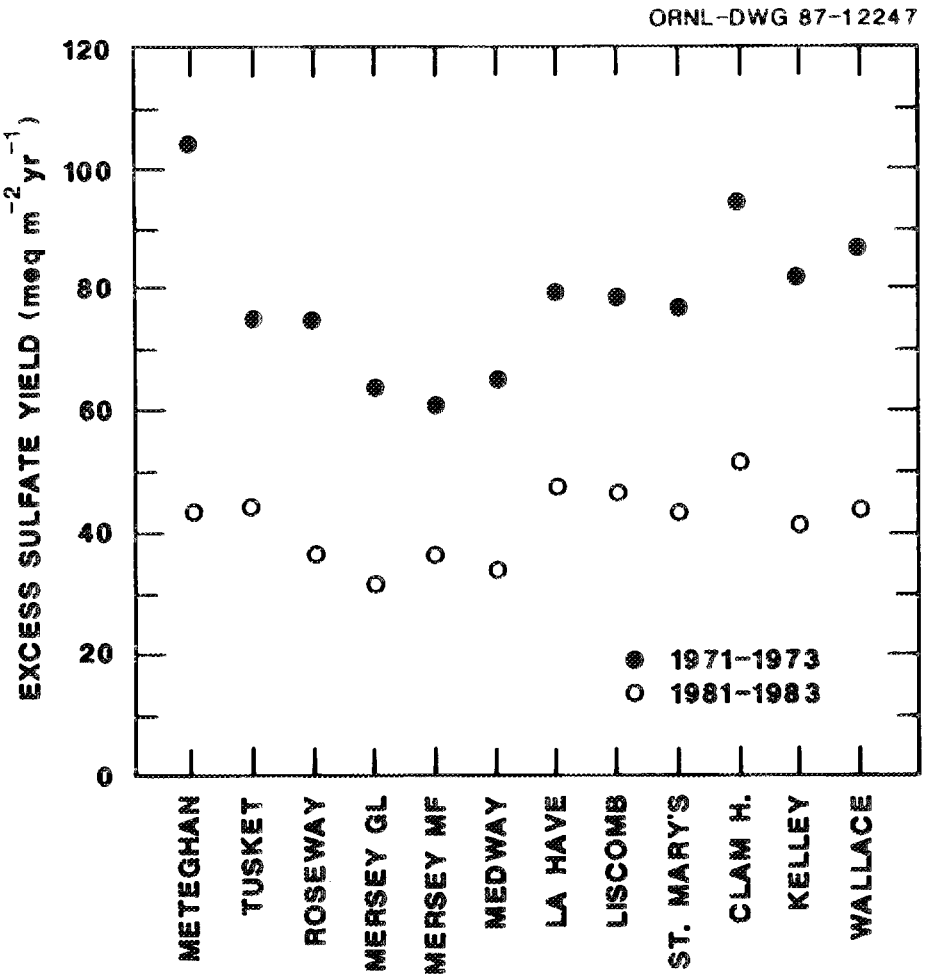

Fig. 6.1. A comparison of excess sulfate yields of 11 rivers in Nova Scotia, 1971-73 (solid circles) and 1981-83 (open circles). (Mersey MF indicates the Mersey River samples at Mill Falls.) Source: Thompson (1986).

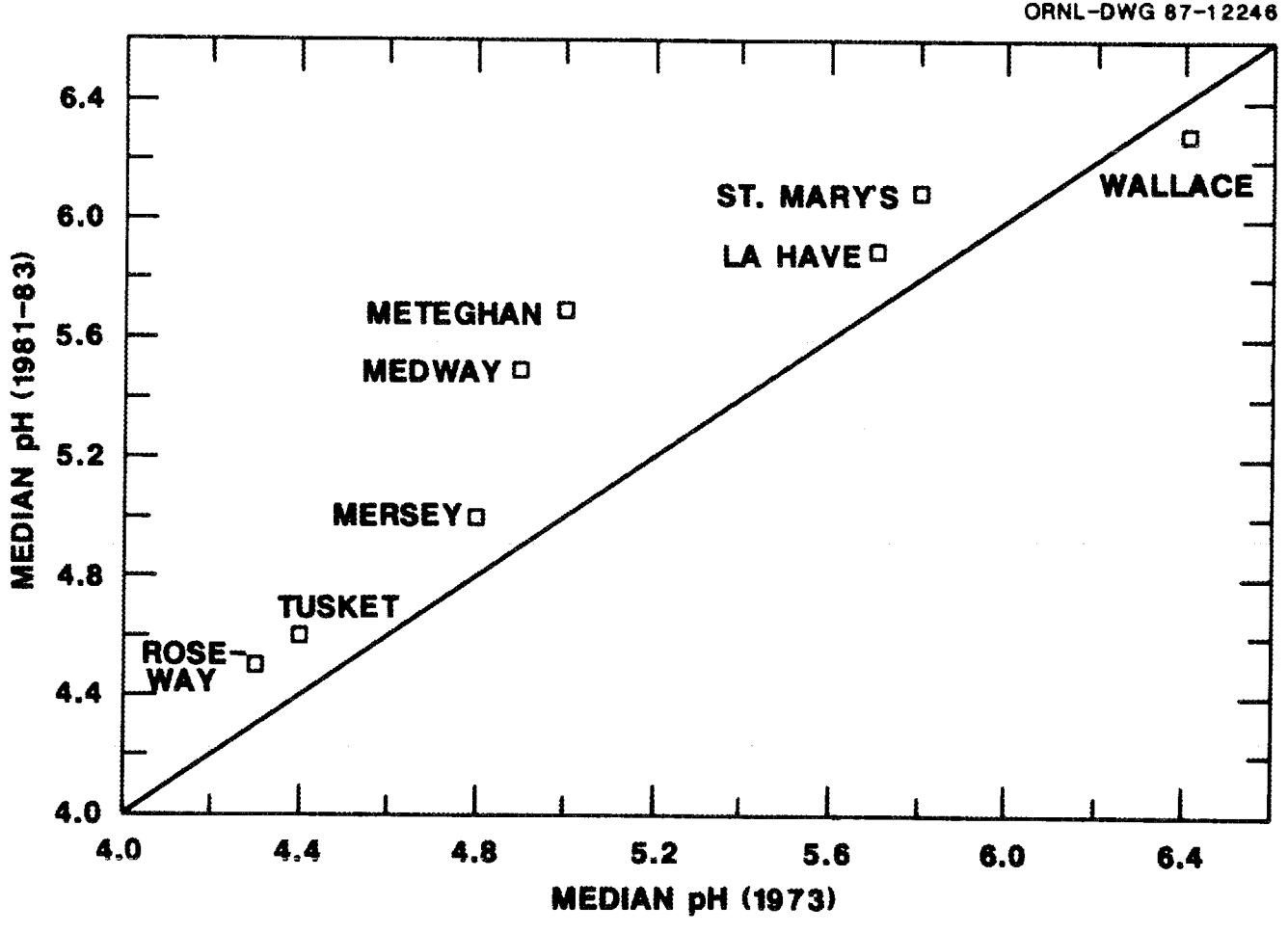

Fig. 6.2. Median pH in 1973 plotted against median pH in 1981-83 for 8 of the 11 Nova Scotia rivers shown in Fig. 6.1. The line represents a 1:1 relationship. Source: Thompson (1982, 1986).

3663-6383 tonne/d from 1970 to 1977; and 1065-2562 tonne/d from 1978 to 1983 (Keller and Pitblado 1986). These declines in emission rates have resulted in a nearly fourfold decrease in average SO_4^{2-} deposition during the summer months from 254 eq ha^{-1} $month^{-1}$ in 1970 to 65 eq ha^{-1} $month^{-1}$ in 1977 (Hutchinson and Havas 1986). Significant trace metal emission declines have accompanied the SO_2 emission reductions.

During the summers of 1982 and 1983, Keller and Pitblado (1986) resampled 209 Sudbury area lakes originally sampled from 1974 to 1976. Between the two study periods, smelter emissions of SO_2 declined by about 50%. Though sampling methods differed somewhat between the two periods, comparative tests indicated that these differences in methods did not cause significant differences in measured parameters. A plot of average lake pH in the 1974-1976 period against average pH recorded from 1981 to 1983 demonstrates that the more acidic lakes (most of which are located relatively near Sudbury) have experienced pH increases (Fig. 6.3). Circumneutral lakes, most of which are relatively far from Sudbury, showed no consistent pattern of change. Reductions in average concentrations of sulfate, nickel, and copper were also greatest in lakes closest to Sudbury. Additionally, other investigators working in the Sudbury area have found that Al, Cu, Ni, and Zn concentrations decreased following reduction in SO_2 emission. These decreases, however, were due to decreased metal emission in the Sudbury area, rather than to changes in pH per se (Dillon et al. 1986).

Keller and Pitblado (1986) performed more detailed analyses for a subset of 21 lakes, each of which had an average pH of <5.5 during the 1974-1976 period. They found statistically significant declines in sulfate and copper concentrations (though not nickel) and statistically significant increases in pH. Significant correlations were found between the decreases in SO_4^{2-}, nickel, and copper concentrations and the distance of these lake from the smelters. On the other hand, watershed-to-lake-area ratios did not correlate significantly with the changes in pH, nickel, or copper and showed only a weak positive correlation with decreases in sulfate concentrations.

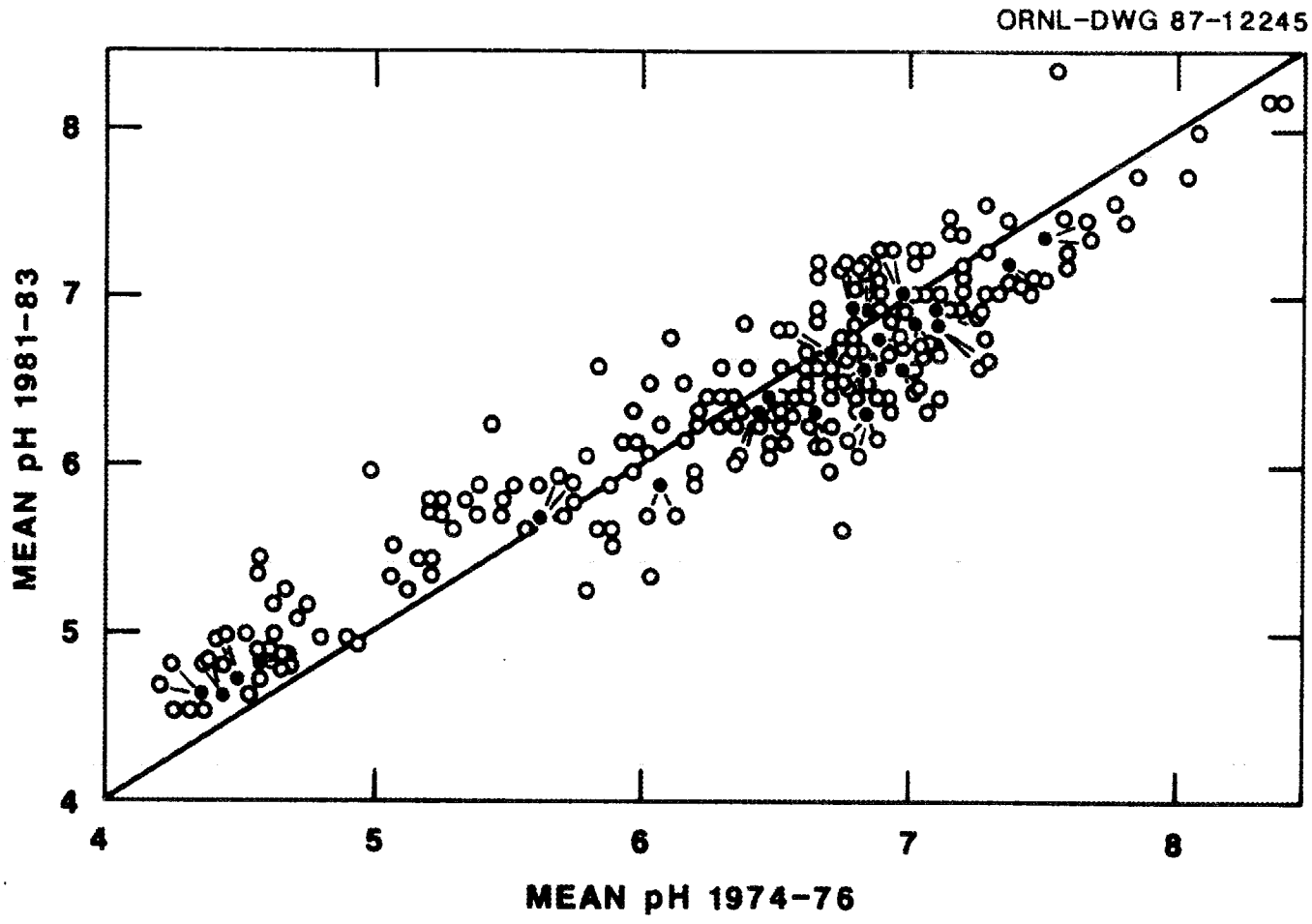

Fig. 6.3. Mean pH in 1974-76 plotted against mean pH in 1981-83 for the Sudbury study lakes. The line represents a 1:1 relationship. Numbers indicate coincident points. Source: Keller and Pitblado (1986).

These findings suggest that deposition levels were more important than watershed factors in determining the rate and extent of surface water chemical responses.

In addition to the surveys of Keller and Pitblado (1986), water chemistry data from five intensively studied lakes in the Sudbury area (Clearwater, Lohi, Middle, Hannah, and Swan) have recently been analyzed by Dillon et al. (1986); the data from Clearwater and Swan are also discussed in LaZerte and Dillon (1984). These data show substantial declines in sulfate in all five lakes during the period from 1978 to 1984, ranging from 25% to 62%. Differences among lakes in the proportional declines were probably due to differences in three factors: distance from Sudbury; lake order (headwater vs second-order lakes); and the extent of anaerobic hypolimnia, which affects bacterial sulfate reduction (LaZerte and Dillon 1984; Dillon et al. 1986). Furthermore, LaZerte and Dillon point out that changes in lake sulfate concentrations are not expected to be directly proportional to the change in local emissions, because some fraction of the area's deposition originates outside the region.

Three of the five lakes were limed; therefore, the changes in these three lakes cannot be attributed to changes in atmospheric deposition. In the other two lakes, mean pH levels increased from 4.23 (1973-77 mean) to 4.57 (1984) in Clearwater and from 3.96 (1977) to 4.80 (1982) in Swan.

The H^+ concentrations in Clearwater declined by over 50%, decreasing from an average of 59 $\mu eq\ L^{-1}$ in 1973-1977 to 25 $\mu eq\ L^{-1}$ in 1983. During this period, sulfate concentrations decreased from an average of 545 $\mu eq\ L^{-1}$ in 1973-1977 to 379 $\mu eq\ L^{-1}$ in 1983 and essentially paralleled declines in lake water H^+ and decreases in emissions of SO_2 from smelting activity in the Sudbury region (Fig. 6.4).

Swan Lake, which was studied less intensively during the same period, showed even more dramatic shifts in H^+ and SO_4^{2-} concentrations; H^+ activity declined 85% (from pH 3.96 to 4.80)

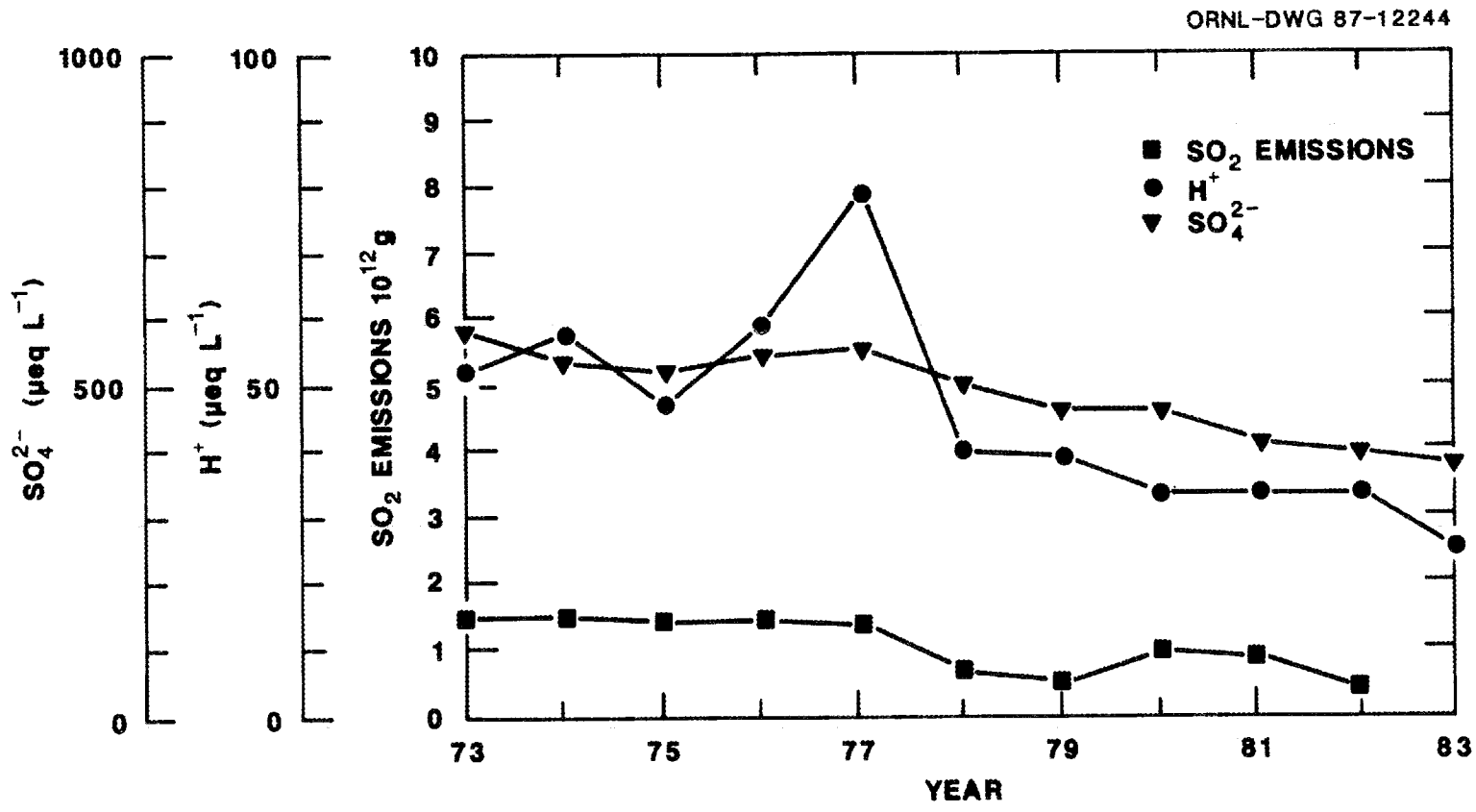

Fig. 6.4. Sulfur emissions at Sudbury (10^{12} g $year^{-1}$) and average H^+ and SO_4^{2-} concentrations in Clearwater Lake (μeq L^{-1}), 1973-83. Data from LaZerte and Dillon (1984).

between 1977 and 1983, and SO_4^{2-} concentrations decreased by 62% from 580 to 220 μeq L^{-1}. LaZerte and Dillon (1984) suggested that the differential in recovery rates shown by the two lakes may reflect differences in hydraulic flushing rates. The hydraulic residence time of Swan Lake is only about 1 year compared to approximately 3 to 4 years for Clearwater Lake.

The data from Clearwater and Swan lakes can be used to examine the two hypotheses mentioned at the start of this section. Swan Lake is the only one of the five lakes not to have received substantial quantities of road salt, therefore permitting an estimate of the F-factor, the ratio of the change in (Ca + Mg) to the change in SO_4^{2-} (Henriksen 1982a). Dillon et al. (1986) estimated F = 0.64 for Swan Lake, a value higher than that reported by Henriksen (1982a) for other locations and indicative of substantial changes in cation export (or substantial transport of cations from the water to the sediment) associated with changes in deposition. As discussed in Sect. 5.1.2.2, Swan Lake had unusually high base cation concentrations for an acidic lake, and the estimated value for F is likely also to be at the upper end of the range for such systems.

LaZerte and Dillon (1984) used the results from Clearwater Lake to refute the hypothesis of Krug and Frink (1983), who contend that a reduction in strong acid deposition will induce the release of equivalent amounts of organic acids with subsequently little change in pH. According to the Krug and Frink hypothesis, reductions in H^+ and SO_4^{2-} deposition in Sudbury should have been accompanied by increases in organic acid concentrations rather than by an increase in pH. No changes in the organic acid content of either Clearwater or Swan Lake were observed, although both lakes experienced substantial increases in pH.

In Baby Lake, a 12 ha lake near Sudbury in a watershed dominated by exposed granitic and gneissic bedrock, SO_4^{2-} concentrations have declined by 50%, and pH has increased from 4.05 in 1972 to 5.8 in 1984 as a result of decreased emissions from smelting activities (Hutchinson and Havas 1986). Conductivity levels have also declined correspondingly.

Similar results have been observed for nearby Alice Lake (Hutchinson and Havas 1986), although the increases in pH for this lake are not as striking as those for Baby Lake. The slower rate of pH recovery reflects the comparatively better buffering of Alice Lake. This is because the Alice Lake watershed is dominated by granitic till, whereas Baby Lake lies in a watershed of exposed bedrock. Also, the pH recovery of Alice Lake may partially reflect artificially high ANC inputs from a limed slag pond.

These empirical studies illustrate several phenomena that must be considered in the context of lake recovery from acidification. A long time series of consistent measurements of both deposition and surface water chemistry is essential. In the case of Sudbury, the variable driving observed changes in surface water chemistry appears to be reductions in atmospheric loadings of sulfate. Measurements of nitrate deposition in the Sudbury area began in 1978 (M. Lusis, Ontario Ministry of Environment, personal communication to D. Marmorek, 1987), after the major decline in sulfate deposition (Fig. 6.4). Therefore, changes in nitrate deposition cannot be entirely discounted as a possible factor affecting lake chemistry, although there were no changes in local emissions of nitric oxides during the period 1977 to 1978 (Hutchinson and Havas 1986). After 1978, there is no observable trend in nitrate concentrations in precipitation or in nitrate loading rates from wet deposition (Ontario Ministry of Environment, 1986), indicating that the declines in lake water pH during this period were not the result of changes in nitrate deposition.

These data raise the issue of the relative effectiveness of controlling SO_2 emissions vs controlling NO_x emissions. Both of the emitted gases are subject to biogeochemical reduction or consumption in natural waters; the net result of such reactions is, in general, the production of an equivalent amount of ANC (Cook et al. 1986; Rudd et al. 1986; Schindler et al. 1986). Atmospheric deposition of NO_x to surface waters is almost completely consumed (>95%) in all

but the most extreme examples of NO_3^- loading (e.g., the Adirondacks; Driscoll and Newton 1985); SO_4^{2-} retention in watersheds on the Canadian Shield and losses due to bacterial SO_4^{2-} reduction typically are less than NO_3^- uptake and in some cases are minimal (Rudd et al. 1986; Kelly et al. 1987; Cook et al. 1987).

In watersheds containing soils with greater sulfate adsorption capacity, such as those in the southeastern U.S., the response in surface water chemistry to decreased deposition is theoretically quite different from the response in systems near Sudbury or elsewhere on the Canadian Shield. Sulfate adsorbed to watershed soil will desorb, delaying the recovery of surface water ANC.

In most acidification models, weathering rates have a major influence on rates of acidification and recovery, although difficulties in empirically estimating weathering rates (Church and Turner 1986; FPRMCC 1986) greatly constrain the level of confidence that can be placed on long-term predictions of the rate of change in surface water chemistry.

6.2.2 Evidence for Biological Recovery from Deposition Reductions

Although water quality has improved in Sudbury-area lakes, many lakes still have a chemistry that is unsuitable for most fish species [e.g., 10% have a pH <5.0; 16% have a pH <5.5 (Keller and Pitblado 1986)]. Information on fish communities in the affected lakes is scarce, although preliminary evidence of improved natural recruitment has been found in several lakes (Keller and Pitblado 1986; Beggs and Gunn 1986).

Yan (1985a) examined the zooplankton community of Clearwater Lake, based on intensive sampling during the period from 1973 to 1984. He computed 11 zooplankton community parameters that have been empirically related to lake water pH or metal levels in published surveys and assessed whether these parameters had changed significantly over time. There were no significant changes in any of the 11 parameters, indicating that the community is not yet recovering. Analysis of possible reasons for the lack of recovery showed that the only feasible explanation is continuing toxic concentrations of hydrogen ion, copper, and nickel.

MacIsaac (1986) and MacIsaac et al. (1986) analyzed data on the rotifer community of Swan Lake and concluded that although some acid-tolerant species have been replaced, the species composition of the rotifer community has not approached that observed in nonacidic lakes.

In conclusion, the preliminary data available suggest that biological recovery is beginning in some lakes in the Sudbury area, but that the original structure, function, and species assemblages have not yet been restored.

6.2.3 Biological Recovery Following Chemical Restoration

Although predictions can be made with some accuracy for both the rate and direction of change in water chemistry associated with restoration procedures, we are currently much less able to predict the resulting biological changes. Nevertheless, studies in chemically restored systems in eastern Canada have demonstrated biological responses. For example, lakes and streams in the Sudbury area, in other areas of Ontario, and in Nova Scotia have been limed. The results of these mitigation efforts are described in detail in FPRMCC (1986) and are summarized in the following sections.

6.2.3.1 The Sudbury Lakes Environmental Study

Between 1973 and 1975 three highly acidic, metal-contaminated lakes near Sudbury, Ontario, were limed by the Ontario government with a combination of calcium hydroxide and calcite, in an attempt to restore fish (Yan and Dillon 1984).

Copper and nickel levels in lake waters were extremely high prior to liming because of metal deposition from nearby smelting operations. Although liming resulted in major increases in pH (to pH >7) and reductions in metal levels, the waters remained highly toxic as a result of residual metals (Yan and Dillon 1984). Introductions of smallmouth bass, Iowa darter, brook stickleback, brook trout, and rainbow trout were all unsuccessful because of this metal toxicity. As inputs of acid and metals from atmospheric deposition continued, the lakes began to reacidify and metal levels increased to their pre-liming levels.

Two of the lakes were fertilized with several annual additions of phosphate after liming (Yan and Lafrance 1984). Although fertilization of limed lakes showed some promise as a tool for delaying reacidification in the Sudbury study, several problems were apparent:

1. concentrations of labile forms of metals were unaffected by nutrient additions;

2. additions of NH_4NO_3 led to pH decreases because of the preferential uptake of NH_4^+ by algae; and

3. phytoplankton systems had higher rates of primary production in response to fertilization than did those in circumneutral waters of similar trophic status, perhaps because higher trophic levels were not present to limit algal biomass in the limed lakes (Yan and Lafrance 1984; Marmorek 1984).

Yan (1985b) compared the zooplankton communities of two limed lakes with 15 nonacidic reference lakes from the Muskoka-Haliburton region. In spite of having circumneutral pH levels for close to a decade, fundamental qualities of the community (including total zooplankton biomass, species richness, mean organism size, relative daphnid biomass, relative cyclopoid biomass, relative predator biomass, and the coefficient of variation in total biomass) differ significantly from those of the reference lakes. Possible reasons for the lack of recovery of the plankton communities in the limed lakes are the absence of fish and invertebrate predators, as well as high copper and nickel concentrations.

In conclusion, lake liming in the Sudbury area has not been successful in restoring the original lake ecosystem function, structure, and community composition. Further experimental work is required to determine whether biological manipulations (i.e., the reintroduction of key species) are necessary.

6.2.3.2 Experimental Lake Neutralization in Ontario

In 1981, the Ontario Ministries of Natural Resources and of Environment initiated a 5-year program to evaluate lake liming as a method to rehabilitate acidified lakes and to protect the biological communities in acid-stressed lakes. Preliminary findings, published by

Booth et al. (1986), showed that in a previously acidified lake, liming sufficiently improved whole-lake water quality so that lake trout could be successfully restocked.

Bowland Lake, an acidic lake near Sudbury that had formerly supported a viable lake trout population, was neutralized in 1983 (Booth et al. 1986). Calcite additions to Bowland Lake raised the whole-lake pH from 5.0 to 6.8 and resulted in reduced concentrations of total aluminum (from 130 to 65 $\mu g\ L^{-1}$). Young lake trout (aged 0+ and 1+) were then restocked; they not only survived, but exhibited good growth. Transferred adult trout spawned in both years after liming, and resident yellow perch population showed both increased growth (older cohorts) and decreased growth (younger cohorts) and a fourfold increase in number. The authors suggested that the yellow perch population responses were possibly due to reduced stress in the older age classes and changes in food supply for the younger age classes. Both were potentially the result of adding calcite to the lake.

The long-term response of Bowland Lake fish populations has yet to be determined, but chemical models suggest that, if it is not relimed, the lake will reacidify to preneutralization water quality by 1990 (FPRMCC 1986). The lake trout population may be in jeopardy much earlier, though. Booth et al. (1986) reported that whole-lake liming did not prevent spring episodes in a nearshore zone under the ice. Caged fish bioassays by these authors showed that the elevated acid and aluminum concentrations in the spawning zone were toxic to hatchery-reared lake trout fry.

After liming Bowland Lake, another low ANC lake was limed in 1984. Unlike Bowland Lake, Trout Lake (near Parry Sound, Ontario) still supported a complex fish community consisting of over 16 species, including lake trout and smallmouth bass. Following liming, no immediate response was observed in the fish community. This was not surprising, since the community had not appeared stressed prior to lake neutralization. The next spring, however, Booth et al. (1986) did observe 100% mortality of caged lake trout fingerlings at one nearshore

site during spring melt, suggesting whole-lake liming was also not entirely effective in mitigating episodic pulses of acidic snowmelt in this lake.

Because whole-lake neutralization does not always protect critical spawning habitat, these investigators also experimented with shoal liming (the addition of limestone gravel to spawning shoals) as an alternative or addition to whole-lake liming. In preliminary results from work at Laundrie Lake, Booth et al. (1986) reported that treating the littoral zone with fine calcite gravel appeared to improve hatching success in lake trout eggs.

6.2.3.3 Liming Nova Scotia Salmon Rivers

Extinction of Atlantic salmon (*Salmo salar*) stocks in 13 Nova Scotia rivers and severe declines in the additional 18 rivers have been attributed to declining pH (Watt 1981; Watt et al. 1983; Watt 1986). To test the feasibility of mitigating salmon losses due to acidification, the Canadian Department of Fisheries and Oceans began experiments in 1981 to add limestone or other substances to streams and lakes (Watt et al. 1984; White et al. 1984). To date, two liming methods have been extensively tested: instream limestone gravel additions (at six river treatment sites) and headwater lake liming (in five lakes). From this work, estimates have been made of the relative costs and effectiveness.

Three years of observations at six sites where limestone gravel had been placed in the streambed proved that under certain conditions, the pH of the streams could be adjusted to >pH 5.0 (Watt 1986). However, low water temperatures during winter and early spring reduced the rate of limestone dissolution, and, at high flows, the amount of limestone gravel that would theoretically be required to give a pH increase of as little as 0.3 units was so large as to be prohibitive. Thus, the overall results were disappointing, leading to the conclusion that this approach is impractical (Watt 1986).

The second approach involved liming headwater lakes to create a reservoir of treated water, which then flowed out by natural discharge to protect downstream salmon habitat. The limestone was usually added

to the lakes as a slurry sprayed evenly over the entire surface. In a four-lake study, Watt et al. (1984) showed that, even with lakes having mean residence times as short as five months, satisfactory lake pH levels could be obtained for up to one year if the limestone dosage exceeded three times the whole-lake acidity. The overdosing provided an excess of lime that settled to the bottom, thus effectively sealing off acid demand from the sediments and dissolving slowly to provide additional ANC at each fall and spring turnover. In those lakes having mean residence times of less than one year, the treatment would have to be repeated annually.

A major problem with the headwater lake liming approach, as noted by White et al. (1984), is that during winter rains the lakes can become covered by a thin surface layer of highly acidic rainwater that flows out of the lake to deliver a downstream acidic shock. Watt (1986) reported that this problem has now been overcome by spreading a layer of dry limestone powder evenly over the ice surface. This form of liming was effective in preventing acidic episodes downstream, because the first water entering the lake from a winter rainstorm is drainage from the ice cover.

With both the instream-gravel and headwater-lake liming methods, positive responses were obtained from Atlantic salmon (Watt et al. 1984). Significantly higher densities of juvenile salmon were found in the immediate vicinity of instream limestone gravel deposits, but this beneficial effect did not persist downstream. In the lake liming studies, salmon parr introduced into the previously barren and toxic outlet stream of Sandy Lake were still surviving one year after liming. In addition, wild native salmon adults migrated (apparently attracted by the higher pHs) into the previously unused outlet stream and spawned successfully. The resulting wild salmon fry showed good survival one year after liming (the end of the reported observation period).

A large area of Nova Scotia has already been rendered barren of Atlantic salmon (Watt et al. 1983). After the acidic deposition problem is brought under control, it will be necessary to mount a

salmon restoration program for the barren habitats. Restoration would be easier and more likely to succeed if a number of native wild local stocks were available to provide a genetically diverse selection of potential donor stocks. To achieve such a gene bank, Watt (1986) recommended that a small number (three to six) of the currently threatened stocks in the Southern Upland of Nova Scotia be preserved by liming headwater lakes to create deacidified refugia in the tributary waters.

Estimating the costs associated with liming lakes in eastern Canada and rivers in Nova Scotia to mitigate effects of acidic deposition is beyond the scope of this assessment. Because of the large numbers of lakes in eastern Canada that are acidic (about 10,000 have ANC <0 $\mu eq\ L^{-1}$; Kelso et al. 1986a), lake liming would obviously be expensive. Recent estimates indicate that the costs would be many tens of millions of dollars (Canadian) (Minns and Kelso 1986).

6.2.4 Chemical and Biological Recovery in Lake 223

The recovery phase of the Lake 223 experiment allows the investigation of chemical and biological recovery in a controlled whole-lake setting (Schindler 1986). Lake 223 in the Experimental Lakes Area received an average of about 9 kg SO_4^{2-} ha^{-1} $year^{-1}$ of wet deposition during the period 1970 to 1982 (Linsey et al., in press). After 2 years of background study in 1974-75, direct experimental additions of sulfuric acid were used to lower the pH of the lake from natural values of >6.5 to 5.0 to 5.1 between 1976 and 1981 (Cook et al. 1986). The pH was kept at 5.0-5.1 during 1981-83 (Schindler 1986). Biological results of the acidification phase of this study are summarized in Sect. 4.3 and in Schindler et al. (1985).

In 1984, sulfuric acid additions were decreased sufficiently to allow the lake to recover to pH 5.4, and in 1985 the pH was allowed to increase to 5.5 to 5.6 (Schindler 1986). As a result of the decrease in sulfuric acid loading, the SO_4^{2-} concentrations decreased in two years from 228 to 202 $\mu eq\ L^{-1}$, a drop of 13%. The decrease in SO_4^{2-} would have been larger, but 1985 was a wet year and acid

additions had to be increased in that year to prevent the pH from recovering more rapidly than planned. ANC increased by 14 μeq L^{-1} as a result of a decrease in base cations of 12 μeq L^{-1} and a SO_4^{2-} decrease of 26 μeq L^{-1} (Schindler 1986).

Phytoplankton composition and diversity in Lake 223 at pH 5.6 during recovery were similar to those observed during the acidification phase of the experiment (Schindler 1986). Zooplankters Holopedium quibberum and Daphnia catawba decreased, perhaps because of the return of planktiverous fishes.

Pearl dace (Semotilus margarita) and white sucker (Catostomus commersoni) began spawning when the pH increased from 5.0 to 5.4, a pH value at which both species reproduced during the acidification phase. Lake trout (Salvelinus namaycush) recovered from their poor condition at pH 5.0 but did not have a successful recruitment at pH 5.4 in 1984.

As expected, species that were eliminated at pH values below 5.7 during the acidification phase have not returned to Lake 223 as of 1985, when pH values increased to 5.5 to 5.6. These species include crayfish (Orconectes virilis), fathead minnows (Pimephales promelas), sculpins (Cottus cognatus), and shrimp (Mysis relicta). These three taxa, which have relatively low ability for recolonization, may have to be restocked when the pH is favorable.

One of the initial findings of the Lake 223 recovery is that species eliminated by acidification may not be able to recover their vacated niches from species that invaded during their absence. For example, brook sticklebacks (Culaea inconstans), a rare species before and during the acidification, invaded the lake in large numbers, and may have displaced the fathead minnow (Schindler 1986). Thus, although the chemical recovery of the lake seems to occur at a predictable rate, biological recovery to the original structure and function of the Lake 223 ecosystem is not predictable.

6.3 IMPLICATIONS

It is clear from the Sudbury and Nova Scotia studies that declines in emissions and deposition lead to chemical recovery. ANCs and pH levels increase; sulfate (and metal concentrations in the case of Sudbury) decline. There are not enough data to evaluate the extent of biological recovery regionally, though site-specific studies near Sudbury suggest that biological recovery is probably either absent or only very slight in many lakes, most likely because of high trace metal concentrations after deposition decreased.

In contrast, mitigation studies in Ontario and Nova Scotia suggest that in the absence of metal contamination, biological recovery can occur, though the rate and extent are still uncertain. There is a need for continued monitoring of both chemical and biological recovery. Such data will be particularly valuable for evaluating the extent to which lakes and streams exhibit a hysteresis in recovery (i.e., a path of recovery very different from simply the reversal of the path followed during acidification).

With respect to mitigation, the lake and river liming experiences in Ontario and Nova Scotia highlight the problems inherent in using full-scale liming operations as a way to rehabilitate and protect aquatic ecosystems:

1. in highly metal-contaminated systems, liming may not reduce metal concentrations to nontoxic levels;
2. without concomitant reductions in deposition, liming becomes a continuous process;
3. liming may not protect the habitat for critical developmental periods of all fish species;
4. the long-term response of resident fish populations to liming has not yet been assessed, and therefore predictions of the response of fish communities to liming are not yet possible;
5. the full recovery of plankton systems may require the re-introduction of fish.

6.4 SUMMARY

Recovery of acidification-damaged freshwater ecosystems can result from either the reduction of acidic inputs or the introduction of substances that neutralize acidity or increase the rate of ANC production. Although few attempts have been made to state the criteria by which attempts at restoring acidification-damaged freshwaters are to be judged, there is a sizeable and growing literature pertaining to chemical and biological recovery in acidic surface waters. In particular, two key issues have received considerable attention in recent years: (a) what factors influence the long-term (years) rate of recovery, and (b) whether recovery results in the same biological community or functional characteristics that existed prior to acidification.

Evidence collected to date strongly supports the notion that aquatic systems affected by acidification will chemically recover after inputs of acidic substances are reduced. For example, decreased sulfate deposition has coincided with decreased concentrations of sulfate and increased ANC and pH in Nova Scotia rivers. Likewise, pH values in the more acidic Sudbury areas lakes have been found to increase during periods when smelter SO_2 emissions decline. Biologically, however, few of these systems have recovered to the point at which the water quality is suitable to support most fish species and more sensitive zooplankton species. Nevertheless, preliminary evidence showing improved fish recruitment is available for a few previously low-pH lakes in the Sudbury area.

In contrast, the beneficial effects of liming surface waters are less clear. For instance, even though field experiments have shown that the chemistry of lakes in the Sudbury area can be at least temporarily improved, the original ecosystem structure and function have not been restored. This is probably partly due to the fact that even liming whole lakes and streams sometimes fails to protect biota and their critical spawning habitat, particularly during acidic episodes.

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