

**EVALUATION OF THE REGIONAL ACIDIFICATION OF
LAKES IN EASTERN CANADA USING ION RATIOS**

by

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NWRI CONTRIBUTION SERIES # 86-79

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Proceedings of the ECE Workshop on Acidification of Rivers and Lakes,
Grafenau, FRG, April 28-30, 1986

ABSTRACT

A high quality data base containing contemporary water quality information for over 7000 lakes in eastern Canada is used to evaluate chemical evidence for acidification that has occurred in response to atmospheric deposition of sulphate. Distribution statistics for alkalinity (Alk), sulphate, and base cation (Ca + Mg) concentrations are presented for 8 subregions which show that the Maritime provinces contain the highest proportion of acidic lakes even though the highest deposition (and lakewater sulphate concentrations) occur in Ontario and Quebec. Consideration of ion ratios (Alk:Ca + Mg, SO_4 :Ca + Mg, and Alk: SO_4) show that the geographical pattern of acidification is a function of both deposition and terrain sensitivity (defined in terms of the terrain's ability to supply alkalinity and/or base cations to its water systems). The ratios provide evidence that a large area of south-central Ontario and southern Quebec has been acidified. The most dramatic evidence of acidification occurs in southern Nova Scotia and a small part of New Brunswick. Northwestern Ontario and Labrador show little evidence of the acidification effect.

Evaluation of the regional acidification of lakes in eastern Canada using ion ratios, by Dean S. Jeffries

RÉSUMÉ

Une base de données de grande qualité, contenant des renseignements actuels sur la qualité de l'eau de plus de 7000 lacs de l'Est du Canada, sert à évaluer les indices chimiques de l'acidification provoquée par les retombées atmosphériques de sulfate. D'après les statistiques de distribution relatives à l'alcalinité et aux concentrations de sulfate et de cations communs (Ca + Mg) dans 8 sous-régions, il appert que la proportion la plus élevée de lacs acides se situent dans les Maritimes, même si les retombées les plus fortes (et les eaux lacustres ayant les concentrations de sulfate les plus grandes) se rencontrent en Ontario et au Québec. D'après les proportions ioniques (alc. : Ca + Mg, SO_4 : Ca + Mg et alc. : SO_4), l'acidification se fait selon une distribution géographique dépendant à la fois des retombées et de la sensibilité du terrain (définie par la capacité du terrain de fournir aux réseaux aquatiques l'alcalinité et/ou les cations communs). Les proportions observées indiquent l'acidification d'une vaste portion du Centre-sud de l'Ontario et du Sud du Québec. C'est dans le Sud de la Nouvelle-Écosse et dans une petite partie du Nouveau-Brunswick que les signes d'acidification sont le plus frappant. Dans le Nord-ouest de l'Ontario et au Labrador, on a observé peu d'effets de l'acidification.

Évaluation de l'acidification régionale dans les lacs de l'Est du Canada d'après les proportions ioniques, par Dean S. Jeffries.

EXECUTIVE SUMMARY

The ionic composition of lakewaters changes in response to acidic deposition. The alteration is usually manifested as decreased acid neutralizing capacity (ie alkalinity), and increased sulphate (and sometimes calcium + magnesium) concentrations. Given the predictable nature of these changes, it is possible to evaluate the geographic extent of the influence of acidic deposition by calculating appropriate ion ratios for lakes spread across eastern Canada.

This report presents the results of such an evaluation in which 3 ion ratios (alkalinity : calcium + magnesium, sulphate : calcium + magnesium, and alkaninity : sulphate) were calculated for a large, high quality data base containing information for over 7000 lakes across all regions of eastern Canada. Contour maps illustrating the variability in the ratio values were prepared.

The ion ratios show that the geographical pattern of lake acidification is a function of both the intensity of atmospheric deposition and terrain sensitivity (defined in terms of the terrain's ability to supply base cations to its surface water systems). Large portions of southern Ontario and Quebec have been acidified although most areas are not yet dominated by acidic lakes. The most dramatic evidence of acidification occurs on southern Nova Scotia and a small part of New Brunswick. Northwestern Ontario and Labrador show little evidence of the acidification effect.

Evaluation of the regional acidification of lakes in eastern Canada using ion ratios, by Dean S. Jeffries

MANAGEMENT PERSPECTIVE

The vast geographic extent of lake acidification occurring in eastern Canada in response to acidic deposition is demonstrated convincingly by consideration of the accompanying changes in lake chemistry. Calculation and presentation of specific ion ratios show that large areas of southern Ontario and Quebec, and smaller areas of Nova Scotia and New Brunswick have been acidified. Only areas experiencing low levels of deposition such as northwestern Ontario and Labrador show little evidence of acidification. Information of this type strengthens the case that major ecological effects due to "acid rain" have already occurred, and will be invaluable during continued negotiations with the US for reduction of sulphate emissions.

This report was prepared in support of a presentation made at a workshop organized by the Economic Commission of Europe entitled "Evidence for the Acidification of Rivers and Lakes", Grafenau, FRG, April 28-30, 1986.

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PERSPECTIVE GESTION

La vaste étendue géographique de l'acidification des lacs de l'Est du Canada par les retombées acides est démontrée de façon convaincante par l'observation des modifications résultantes de leur chimie. Le résultat du calcul des proportions de certains ions montre que de vastes portions du Sud de l'Ontario et du Québec et des zones moins grandes de la Nouvelle-Écosse et du Nouveau-Brunswick sont acidifiées. Seules les zones où les retombées sont rares - par exemple, le Nord-ouest de l'Ontario et le Labrador - laissent voir peu de signes d'acidification. Ce genre de renseignements appuie la thèse voulant que des effets écologiques majeurs dus aux "pluies acides" se soient déjà fait sentir; ils seront d'une valeur inestimable pour la suite des négociations avec les É.-U. en ce qui a trait aux émissions de sulfate.

Le présent rapport a été préparé pour appuyer un exposé présenté à un atelier organisé par la Commission économique pour l'Europe, intitulé "Evidence for the Acidification of Rivers and Lakes", à Grafenau en RFA, les 28-30 avril 1986.

Évaluation de l'acidification régionale des lacs de l'Est du Canada d'après les proportions ioniques, par Dean S. Jeffries.

INTRODUCTION

The relationship between acidification of aquatic ecosystems and atmospheric deposition has been extensively analyzed, reported, and summarized (Drablos and Tollan, 1980; NAS, 1981; US-Canada, 1983). Literally hundreds of site specific studies have concluded that acidic deposition is instrumental in effecting decreases in surface water pH and/or alkalinity. However, evaluation of the "acidification status" of lakes on a sub-continental scale in eastern Canada has received lesser attention due to the paucity of good quality historical data, lack of coordination of the many research and monitoring agencies, and the massive data handling problems that such a project entails.

Harvey et al. (1981) and US-Canada (1983) have used specific, geographically limited data sets to show the variation that exists in surface water quality across eastern Canada. Variation in the ionic composition of these waters, particularly increased sulphate and/or decreased alkalinity concentrations, was evaluated in relation to the geographical variation in atmospheric deposition. Results were usually presented in the form of maps of individual ion concentrations, often in conjunction with terrain sensitivity maps based on soil and bedrock geochemistry. Interpretation of such single ion maps is inherently limited by the large number of confounding climatic, biological, and geochemical factors which can influence the individual concentrations. This is particularly true for alkalinity.

Wright (1983) successfully used similar data to refine Henriksen's empirical "acidification model" for application to North American lakes. The model predicts that lake water sulphate concentration elevated above a background level due to atmospheric deposition, is balanced by either a decrease in alkalinity or an increase in base cation (Ca + Mg) concentration. The inter-relatedness of these parameters suggests that consideration of ion ratios such as Alk:Ca + Mg , $\text{SO}_4\text{:Ca + Mg}$, or Alk:SO_4 will be a superior method for assessing the acidification status. This is particularly true for eastern Canada where the vast area of potentially sensitive terrain (ie having silicate bedrock) possesses a wide diversity of climatic and geochemical characteristics which are manifested in large variances in basic cation and alkalinity concentrations.

Jeffries et al. (in press) recently compiled a large, high quality, computerized data base which lends itself to an evaluation using ion ratios. This paper presents the early results and interpretation of this work.

CHARACTERISTICS OF ION RATIOS

Wright's acidification equation shows the dynamic inter-relationship which exists among SO_4 concentration (which is itself a function of deposition intensity), Alk, and Ca + Mg concentrations. It should be noted that the concentrations referred to here (as in Wright's work) are all "sea-salt" corrected; the reader

should therefore assume that the SO_4 and $\text{Ca} + \text{Mg}$ values discussed and/or presented throughout this paper are likewise corrected, where appropriate. While most studies suggest that loss of Alk is the predominant response to elevated SO_4 , certain locations such as the heavily impacted Sudbury area exhibit high concentrations of $\text{Ca} + \text{Mg}$. In either case however, the Alk: $\text{Ca} + \text{Mg}$ ratio will decrease from a value near unity (for the ideal, clear lake experiencing low deposition) to zero as the alkalinity is exhausted, to negative values as the lake becomes increasingly acidic. The sensitivity of this ratio is dependent on the absolute magnitude of the numerator and denominator; that is, for a constant intensity of SO_4 deposition, the ratio for a lake having relatively high concentrations of Alk and $\text{Ca} + \text{Mg}$ will decrease to a lesser extent than for a lake having low concentrations of these ions. Hence the Alk: $\text{Ca} + \text{Mg}$ ratio not only responds in a predictable manner to differing SO_4 levels, but also the magnitude of the response (ie the ratio sensitivity) directly reflects the terrain's capability of supplying basic cations and Alk - a reasonable definition of "terrain sensitivity".

Naturally occurring organic acids may influence the ratio, most likely by reducing the Alk concentration and thereby decreasing the ratio from the ideal unperturbed value of approximately one. The organic anion concentration (A^-) of waters is easily estimated if dissolved organic carbon (DOC) and pH are known (Oliver et al., 1983). If it can be assumed that the A^- content is independent of lake acidity and reflects an equivalent reduction in Alk, then it is possible to correct the ratio for the influence of A^- . The importance of considering A^- is greatest when terrain (and ratio)

sensitivity is high.

The $\text{SO}_4:\text{Ca} + \text{Mg}$ ratio will ideally approach but not reach zero in areas of background deposition since background SO_4 concentrations are approximately 20 - 40 ueq/L (Wright, 1983; Jeffries et al., in press); similarly, it will approach unity as sulphate replaces alkalinity or A^- in high deposition areas. The ratio can in fact exceed unity when Alk becomes negative. Given that A^- more likely effects a reduction in background Alk rather than an increase in $\text{Ca} + \text{Mg}$, the $\text{SO}_4:\text{Ca} + \text{Mg}$ ratio is relatively less affected by the presence of organic acids. Once again, since $\text{Ca} + \text{Mg}$ is in the denominator of the ratio, the ratio's sensitivity will reflect the terrain's ability to supply base cations to its water systems.

The $\text{Alk}:\text{SO}_4$ ratio provides a direct indication of the SO_4 for Alk replacement that occurs during the acidification process. It will range from very large values (>10) when Alk is abundant (ie in relatively insensitive terrain) and SO_4 is low (ie low deposition areas) to values slightly <0 when Alk is negative. Important ratio values are 1.0 occurring when $\text{Alk} = \text{SO}_4$, 0.5 when SO_4 is 2-fold greater than Alk, and 0.2 when SO_4 is 5-fold greater than Alk.

Given the above discussion, perhaps the best overall ratio would be $\text{Alk} - \text{SO}_4 + \text{A}^-:\text{Ca} + \text{Mg}$ which contains both a correction for the organic anion effect (once again assuming that A^- affects Alk only) and also combines the influences of Alk and SO_4 into one

ratio value. Ideally, this ratio would vary from +1 under conditions of background deposition, to zero when Alk and SO_4 are equivalent in a low A^- lake, and to increasingly negative values as SO_4 becomes the dominant anion and Alk becomes negative. Clearly, all of the above ratios can be strongly influenced by the existence of a localized geological source of 1 or more of the components (eg localized limestone, dolomite, or sulphide deposits). Such situations do exist in eastern Canada and were observed during the data analysis below.

DATA DESCRIPTION AND PROCESSING

The data base used for this evaluation is similar to that compiled by Jeffries et al. (in press) except that substantial additions have been made for Nova Scotia and Quebec by including the data from Underwood (in press) and Dupont and Grimard (in press) respectively. A full description of the data base characteristics is presented in Jeffries et al. (in press) and therefore will not be repeated here. Briefly, it is a compilation of several smaller federal and provincial government data sets and contains information for approximately 7000 lakes of which 4500 are located in Ontario. Almost all data were collected during or after 1980; hence, this data set reflects the contemporary water quality condition. Most values in the data base reflect average whole-lake composition either through the use of integrated sampling methodologies, or by appropriate mathematical manipulation of non-integrated data. All Alk measurements presented here were obtained using a Gran (or equivalent) procedure, and thus, the data base contains negative values.

Sulphate analyses were conducted by either ion chromatography (or equivalent) or automated colorimetry, usually the methyl thymol blue (=MTB) procedure. IC determined SO_4 was used in preference to MTB values whenever possible; when only MTB data were available, the data were included in the following evaluation only when sample colour was <30 Hazen units and/or DOC was <6 mg/L. The elimination of MTB SO_4 data for highly coloured waters is necessary due to the analytical interferences present for this type of sample (Kerekes et al., 1984).

Calcium and Mg analyses were performed separately by appropriate means, usually AAS. All SO_4 and Ca + Mg data used in the following evaluation have been sea-salt corrected except for the Ontario subset where a lack of chloride data prevented this calculation. Sea-salt influence in Ontario is considered negligible however. Finally, the data base has been screened to remove all samples exhibiting obvious local anthropogenic influences (eg. road salt, acid mine drainage, etc.).

As noted earlier, ratios involving Alk may be influenced by A^- . The data base is incomplete for DOC at this time which has precluded calculating ratios with an A^- correction. Jeffries et al. (in press) showed that for those lakes where calculation of A^- was possible and Ca + Mg concentrations were low (in Nova Scotia for example), there is a relatively even distribution of A^- across all classes of Alk. That is, high A^- were just as likely to occur in lakes with higher Alk as in lakes with low or negative Alk. Thus it

appears that the influence of A^- on the ratios will vary greatly from lake to lake. Recalling that the $SO_4:Ca + Mg$ ratio is probably least affected by A^- , the possible influence of A^- will be assessed by calculating each of the first 3 ratios discussed above and comparing the results.

The Canadian subregions defined by Jeffries et al. (in press) will be used to facilitate the discussion below (see Figure 1). Distribution statistics were calculated for each of the 8 Canadian subregions (ie NW ONT, NE ONT, SC ONT, QUE, LAB, NB, NS, and NF) in order to provide insight into the between region variability of the data. A computer program was written to facilitate visual presentation of the ion ratios. The map of eastern Canada was divided into a 20 X 20 minute latitude and longitude grid and each lake in the data base was appropriately located within its correct grid square. The median ratio value (and median latitude and longitude) within each grid square was determined and the median automatically plotted at the proper location on a computer generated mercator projection of eastern Canada. The number of lakes falling within any given 20 X 20 minute square ranged from 1 to 110. Clearly the median ratio determined for a square containing 1 lake was less reliable than than for a square containing many lakes; thus the map was contoured manually in order to make allowances for this and/or other localized factors.

RESULTS AND DISCUSION

Table 1 gives the 5th, 50th (ie median), and 95th percentile

CANADIAN REGIONS OF INTEREST

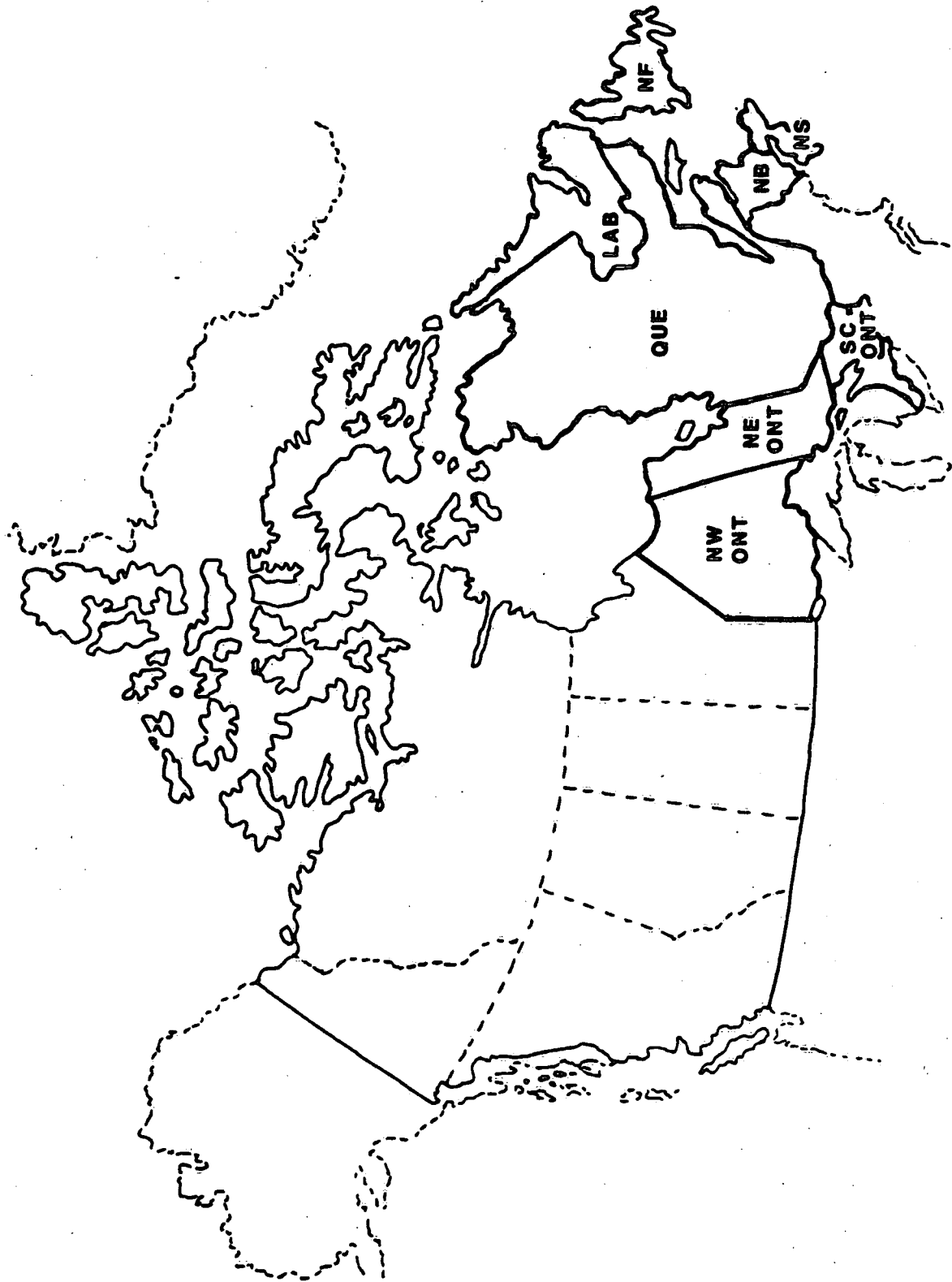


Figure 1 : Subregion designation for eastern Canada (after Jeffries et al., in press)

concentrations (ueq/L) for Alk, and Ca + Mg for the 8 Canadian subregions. Alk and Ca + Mg concentrations are generally greatest in Ontario due to the presence of small amounts of CaCO_3 in the glacial tills of the region. The quantity of CaCO_3 gradually increases northward towards the source area in the Hudson Bay Lowlands (Shilts, 1981) which accounts for the exceptionally high concentrations (for nominally Shield lakes) in the upper tail of the data distributions in Ontario. The negative alkalinity in the low tail of the NE ONT distribution reflects the significant number of acidic lakes occurring near Sudbury. Carbonate material is not present in the tills of most of Quebec and the Maritime provinces, and therefore, Alk and Ca + Mg are consistently lower than in Ontario. Nova Scotia (NS) has the lowest median Alk and Ca + Mg for all the subregions (over 30% of its lakes are currently acidic, $\text{Alk} < 0$, Jeffries et al., in press). Labrador (LAB) is the region furthest downwind from the major source areas in the central part of the continent; its median Alk concentration (46 ueq/L) approximates a reasonable background value.

The distribution of SO_4 among the subregions is also given in Table 1. The greatest median concentrations occur in SC ONT, NE ONT, and QUE where the deposition is also highest. In fact, the region to region pattern of median concentration closely follows the expected level of deposition across all eastern Canada. Hence, background SO_4 (ie in LAB) is approximately 24 ueq/L in agreement with the analysis of Wright (1983). The high SO_4 in the upper tail for NE ONT once again reflects the influence of the major local source at Sudbury.

Table 1. Fifth, fiftieth, and ninety-fifty percentiles of lake water concentrations of alkalinity, sulphate, and Calcium + Magnesium (µeq/L) for subregions of Eastern Canada (see Figure 1). Ratio components are sea-salt corrected were appropriate.

| SUBREGION | n* | ALKALINITY | | | SULPHATE | | | CALCIUM + MAGNESIUM | | |
|-----------|----------------------|------------|----------|------------|----------|----------|-----------|---------------------|-----------|------------|
| | | 5th | 50th | 95th | 5th | 50th | 95th | 5th | 50th | 95th |
| NW ONT | 216-1078 | 46 | 254 | 1738 | 33 | 75 | 121 | 81 | 221 | 2093 |
| NE ONT | 793-1805 | -11 | 165 | 2194 | 33 | 148 | 282 | 143 | 441 | 2623 |
| SC ONT | 750-1578 | 5 | 63 | 1331 | 105 | 158 | 201 | 126 | 199 | 447 |
| QUE LAB | 1268-1452 180-203 | 4 14 | 52 46 | 536 166 | 22 9 | 93 24 | 186 43 | 65 31 | 163 69 | 713 245 |
| NB | 52-81 | - 2 | 38 | 185 | 47 | 77 | 98 | 43 | 90 | 208 |
| NS | 275-413 | -30 | 10 | 122 | 12 | 50 | 106 | 11 | 53 | 191 |
| NF | 176-268 | - 0 | 28 | 244 | 10 | 31 | 55 | 21 | 79 | 461 |

*range in n depending on ion considered.

Table 2 presents the 5th, 50th, and 95th percentile values for 3 ion ratios for the 8 Canadian subregions. The median Alk:Ca + Mg ratios are highest for LAB where deposition is lowest and for NE ONT where the CaCO_3 content of the tills overrides the influence of atmospheric deposition. Lowest median values occur in SC ONT where the CaCO_3 influence is smallest (for Ontario) and the deposition is highest, and in NB and NS where terrain sensitivity is high and deposition is moderate. Only NB, NS, and NF contain a significant number of negative Alk:Ca + Mg ratios in the low tail of their data distributions. Those regions with CaCO_3 in the tills or receiving only low levels of deposition exhibit high tail values near one as predicted earlier.

The SO_4 :Ca + Mg ratio is also strongly influenced by both regional geochemistry and deposition. Hence, lowest median ratios occur for NE ONT and NW ONT due to the high Ca + Mg concentrations, and for LAB due to low SO_4 deposition. Highest median values occur where deposition is high enough to override moderating geochemical conditions (ie SC ONT) or where the terrain is very sensitive (ie Ca + Mg very low in NB and NS). Those SO_4 :Ca + Mg ratios exceeding one in the high tail of the NB, NS, and NF distributions reflect the higher incidence of acidic lakes occurring in these subregions.

The Alk: SO_4 ratios may be interpreted in the same manner, ie high deposition and/or high terrain sensitivity yield low median values while low deposition and/or low terrain sensitivity yield high

Table 2. Fifth, fiftieth, and ninety-fifty percentiles of the ion ratios Alk:Ca+Mg, SO₄:Ca+Mg, and Alk:SO₄ for subregions of Eastern Canada (see Figure 1). Ratio components are sea-salt corrected where appropriate.

| SUBREGION | n* | Alk:Ca+Mg | | | SO ₄ :Ca+Mg | | | Alk:SO ₄ | | |
|-----------|-----------|-----------|------|------|------------------------|------|------|---------------------|------|------|
| | | 5th | 50th | 95th | 5th | 50th | 95th | 5th | 50th | 95th |
| NW ONT | 172-333 | 0.1 | 0.6 | 1.0 | 0.0 | 0.4 | 0.8 | 0.4 | 2.3 | 35.5 |
| NE ONT | 667-792 | 0.0 | 0.7 | 1.1 | 0.0 | 0.2 | 1.1 | -0.0 | 1.6 | 49.2 |
| SC ONT | 723-814 | 0.0 | 0.3 | 0.7 | 0.3 | 0.8 | 1.1 | 0.0 | 0.3 | 2.2 |
| QUE | 1260-1268 | 0.0 | 0.4 | 1.0 | 0.1 | 0.5 | 1.1 | 0.0 | 0.6 | 6.9 |
| LAB | 142-179 | 0.3 | 0.8 | 1.0 | 0.1 | 0.3 | 0.7 | 0.6 | 2.4 | 8.7 |
| NB | 49-56 | -0.0 | 0.3 | 0.8 | 0.3 | 0.9 | 1.6 | -0.0 | 0.3 | 2.3 |
| NS | 234-292 | -2.3 | 0.3 | 0.9 | 0.2 | 0.8 | 2.5 | -0.5 | 0.4 | 3.2 |
| NF | 143-176 | -0.0 | 0.5 | 1.0 | 0.0 | 0.5 | 1.4 | 0.0 | 1.1 | 8.5 |

*range in n depending on ratio calculated.

median values.

Figures 2, 3, and 4 present the geographical distribution of the $\text{Alk:Ca} + \text{Mg}$, $\text{SO}_4:\text{Ca} + \text{Mg}$, and Alk:SO_4 ratios respectively.

The pattern presented by each ratio is remarkably consistent among the 3 Figures. All show large areas of south-central Ontario and southern Quebec as having undergone significant acidification, either in terms of Alk depletion or SO_4 elevation. The influence of localized higher deposition related to point sources (Sudbury and Noranda in NE ONT and QUE respectively) is visible, as is the influence of topographic control over deposition, eg the reflection of higher deposition occurring in response to rapidly increasing elevation to the north of the upper reaches of the St Lawrence River valley.

Geological moderation of the occurrence of acidification is obviously evident in southern Ontario at the contact between the Canadian Shield and the carbonate bedrock to the south, in northern Ontario for the reasons discussed earlier, and even in the narrow band of lower sensitivity terrain along the Ottawa River valley between Quebec and Ontario.

The maps all show that the most dramatic indication of acidification occurs in southern NS and a small part of NB principally due to terrain sensitivity rather than strongly elevated deposition. The fact that both the $\text{SO}_4:\text{Ca} + \text{Mg}$ ratio and the $\text{Alk:Ca} + \text{Mg}$ ratio exhibit much the same pattern gives credence to the conclusion of Jeffries et al. (in press) that A^- can not, on its own, explain the occurrence of many acidic lake systems in this area. The southern portion of the island of Newfoundland exhibits a gradient of

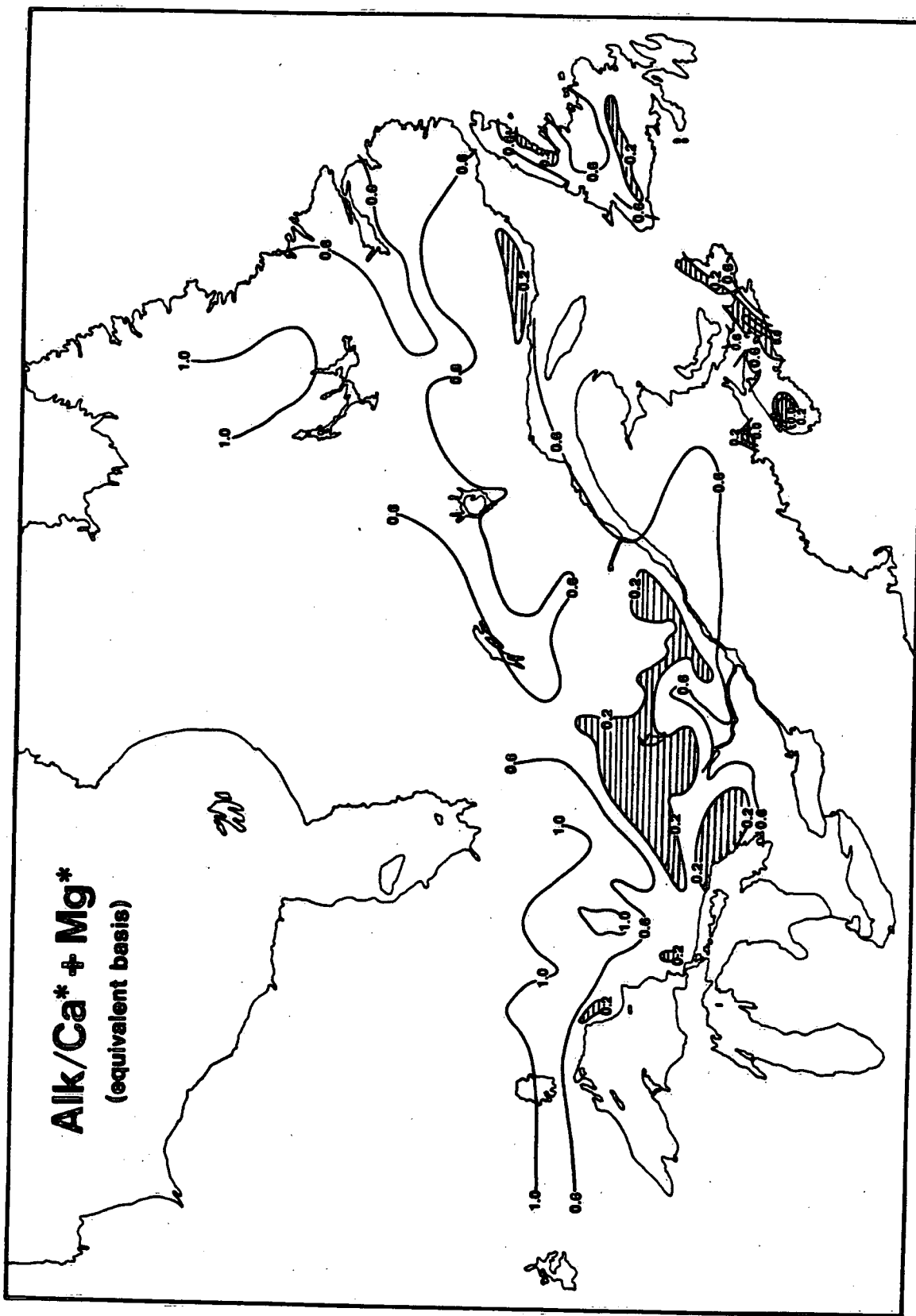


Figure 2 : Geographical distribution of the Alk:Ca + Mg ratio across eastern Canada. Cross-hatched areas exhibit the most pronounced indication of surface water acidification.

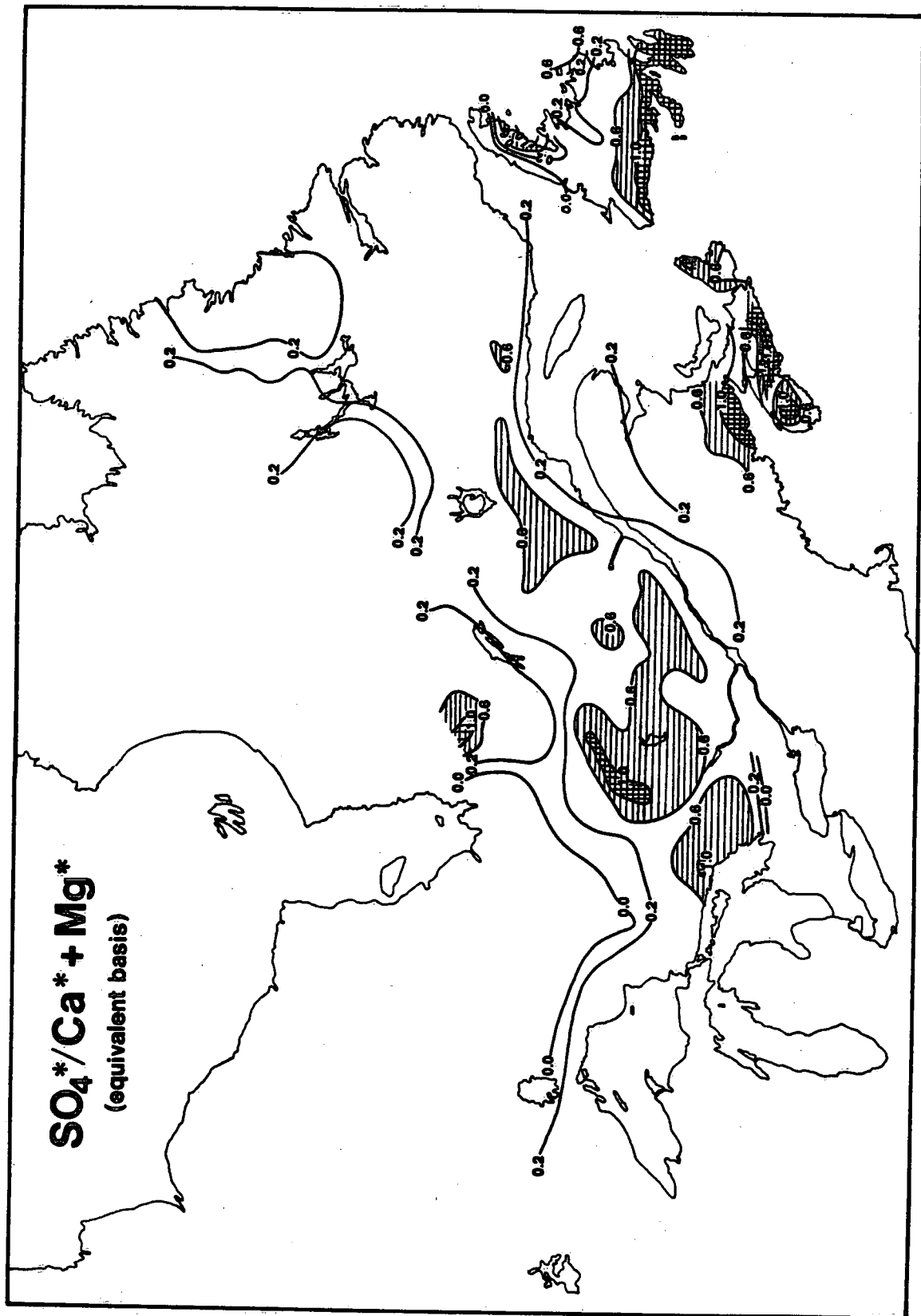


Figure 3 : Geographical distribution of the $\text{SO}_4:\text{Ca} + \text{Mg}$ ratio across eastern Canada. Cross-hatched areas exhibit the most pronounced indication of surface water acidification.

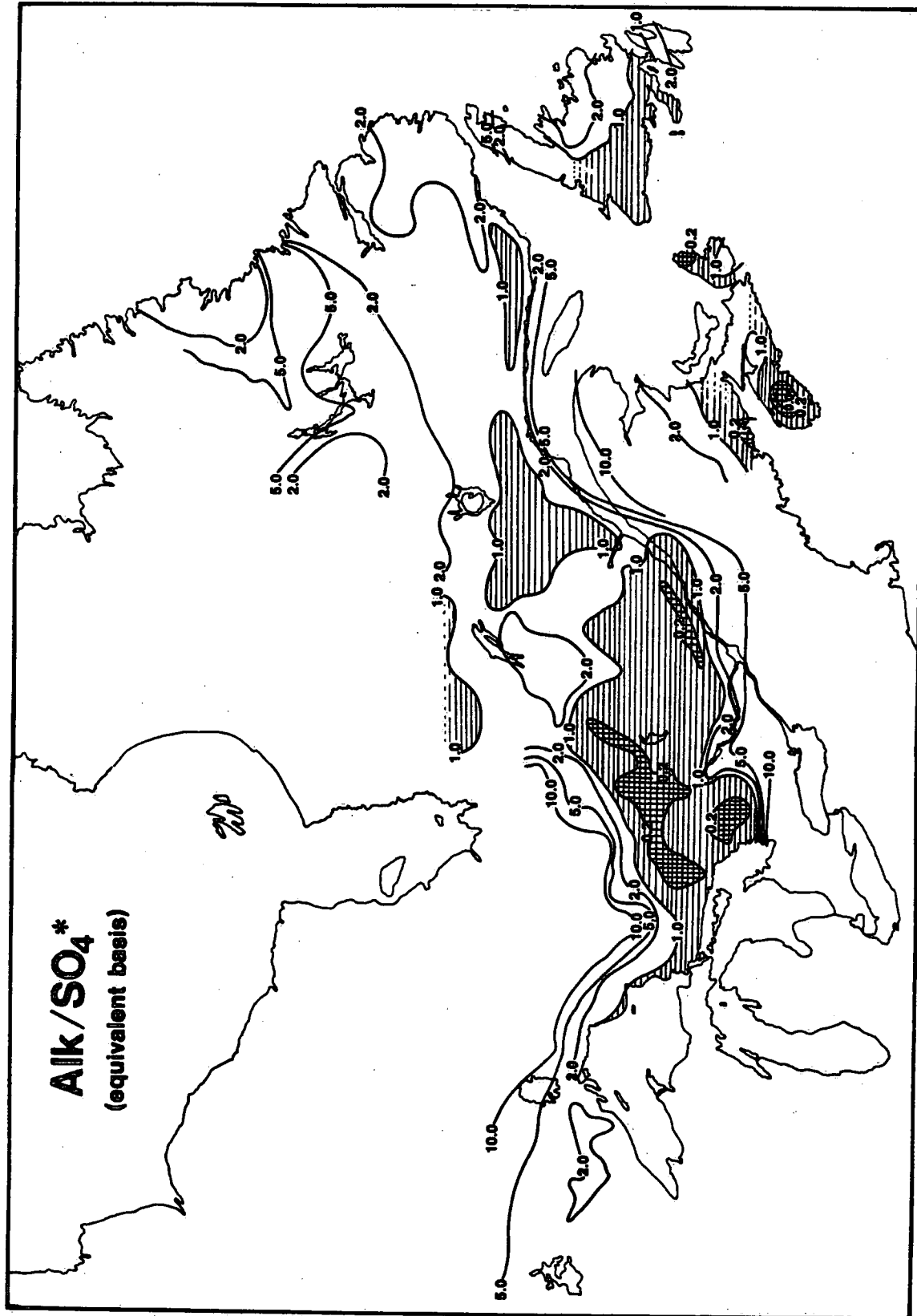


Figure 4 : Geographical distribution of the $\text{Alk}:\text{SO}_4$ ratio across eastern Canada. Cross-hatched areas exhibit the most pronounced indication of surface water acidification.

acidification from highest in the south to lowest in the north, presumably reflecting the similar gradient in deposition. Finally, all the maps show that NW ONT and LAB have experienced lesser acidification to date.

ACKNOWLEDGEMENTS

This paper could not have been produced without the able assistance of the staff of the CCIW Computer Centre. In particular, I wish to thank Bob Duffield and Angelo Zingaro for their help.

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