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**Analysis of Southeastern Canada  
Lake-Water Chemistry Data in  
Relation to Acidic Deposition**

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Environmental Sciences Division  
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ENVIRONMENTAL SCIENCES DIVISION

ANALYSIS OF SOUTHEASTERN CANADA LAKE-WATER CHEMISTRY DATA  
IN RELATION TO ACIDIC DEPOSITION

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## ABSTRACT

Olson, R. J., R. B. Cook, B. M. Ross-Todd, and J. J. Beauchamp. 1991. Analysis of Southeastern Canada Lake-Water Chemistry Data in Relation to Acidic Deposition. ORNL TM-11576. Oak Ridge National Laboratory. Oak Ridge, Tennessee.

Lake-water chemistry data were obtained for lakes in southeastern Canada to study relationships between atmospheric deposition and acid-base chemistry as part of the National Acid Precipitation Assessment Program State of Science and Technology reports. Quality assurance checks were made to ensure that the data used were of sufficient quality and were comparable to data from the United States. Ninety-eight percent of the 8506 sampled lakes had pH, ANC, and  $\text{SO}_4^{2-}$  data and were used in our analyses. Of these, we created a subset of 4017 lakes having data for more variables ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ , DOC, and conductivity) to analyze potential sources of lake-water acidity. The objectives of this work were to determine the geographical extent and number of potentially affected systems and to infer causes of acidification based on ion ratios.

In southeastern Canada, 3041 (38%) of the sampled lakes have an acid neutralizing capacity (ANC)  $\leq 50 \mu\text{eq L}^{-1}$ . Of these, 381 (5% of the sampled lakes) are acidic (ANC  $\leq 0 \mu\text{eq L}^{-1}$ ). The Sudbury/Noranda region has the greatest percentage of sampled acidic lakes (24%), which comprise 43% of the sampled acidic lakes in all of southeastern Canada. In contrast, in the South Atlantic Region 90 of the 764 sampled lakes (12%) and in southeastern Ontario 86 of the 2086 sampled lakes (4%) are acidic. No acidic lakes were sampled in West Ontario. Acidic lakes totaled <1% of the 1382 lakes sampled in North Ontario, a region whose lakes are characterized by generally low atmospheric deposition and high levels of dissolved organic carbon.

Using a subset of 4017 lakes that have a relatively complete set of water chemistry variables, we identified sulfate- vs organic-dominated low-ANC systems. Acidic lakes with concentrations of  $\text{SO}_4^{2-}$  greater than  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  are primarily located in Sudbury/Noranda (106 out of 230 acidic lakes). These lakes have relatively low organic anion concentrations and have experienced acidification as a result of historically high  $\text{SO}_4^{2-}$  deposition originating from local smelters.

Acidic lakes having concentrations of  $\text{SO}_4^{2-}$  greater than  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  are also located in southeast Ontario and in the Maritime Provinces. For these acidic lakes, lake-water  $\text{SO}_4^{2-}$  is much greater than organic anion concentrations, suggesting that these lakes have experienced some acidification as a result of atmospheric deposition. For the acidic lakes in Maritime Provinces having concentrations of  $\text{SO}_4^{2-}$  greater than  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$ , organic anions are present at concentrations similar to but less than those of  $\text{SO}_4^{2-}$ , indicating that a

combination of mineral acids and organic acids are responsible for the current acid-base chemistry.

Twenty-eight acidic lakes ( $ANC \leq 0 \mu\text{eq L}^{-1}$ ) located in the South Atlantic region (primarily Nova Scotia) have concentrations of organic anions greater than  $\text{SO}_4^{2-}$  and greater than  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$ . These proportions of anions suggest that the acidity is substantially influenced by organic acids.

## 1. INTRODUCTION

Lake-water chemistry data were obtained for 8506 lakes in southeastern Canada to study the effects of acidic deposition, especially in comparison to similar work in the United States. Quality assurance (QA) checks were made to ensure that the data used were of sufficient quality and were directly comparable to data from the United States. In addition, we divided the data into subsets to identify lakes having key variables (pH, ANC,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ , DOC, and conductivity) required for the analyses. Using selected subsets, our analysis showed general patterns of low-ANC lakes associated with high-deposition levels similar to results reported by Canadian scientists (Jeffries et al. 1986, Schindler 1988, Neary and Dillon 1988, Neary et al. 1990, Jeffries in press). Sulfate- vs organic-dominated low-ANC systems were identified by the ratio of  $\text{SO}_4^{2-}$  to  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  vs the ratio of organic anion [calculated using the Oliver et al. (1983) model] to  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$ . Sulfate-dominated lakes with low ANC were generally located in areas receiving relatively high  $\text{SO}_4^{2-}$  deposition, whereas organic-dominated lakes with low ANC were located in areas containing higher percentages of peatlands and relatively low levels of  $\text{SO}_4^{2-}$  deposition.

This study (1) compares and contrasts the status and extent of low-ANC lakes in southeastern Canada in relation to the needs of State of Science/Technology Report No. 9 (SOS/T 9, Baker et al. 1990) prepared for the U.S. National Acid Precipitation Assessment Program (NAPAP), by using analysis techniques developed for SOS/T 9 and (2) examines the relationships between atmospheric deposition and the acid-base chemistry of lakes in southeastern Canada. Because many other factors (e.g., hydrology, soil chemistry, and bedrock geology) influence lake-water chemistry and its relationship with acidic deposition, these factors were considered in our analyses whenever the data permitted.

## 2. DATA AND METHODS

### 2.1 Lake-Water Chemistry Data Acquisition and QA Checks

Lake chemistry, lake inventory, and watershed attribute data were obtained for southeastern Canada from provincial and federal government research scientists. Data for Ontario were obtained in December 1988 (B. Neary, Ontario Ministry of the Environment, personal communication, December 1988) and the revised data set for other provinces (Quebec, Newfoundland, Nova Scotia, Prince Edward Island, and New Brunswick) was obtained in June 1989 (D. S. Jeffries, National Water Research Institute, Environment Canada, personal communication, June 1989).

Canadian lakes were sampled for various reasons using different sample designs. Furthermore, the sample designs were not documented in the data base, although a brief description of the sampling design

for some of the surveys was given in Jeffries et al. (1986) and Neary et al. (1990). Data for Ontario were compiled from 31 surveys; Neary et al. (1990) classified the potential biases associated with these surveys as (1) no known bias: 16; (2) possible bias towards large lakes: 9; and (3) possible bias on known geology or fish status: 6. The lakes were sampled at different times of the year during the 1980s; medians were provided to us for lakes having multiple samples. Different analytical and QA/quality control procedures were used, and inconsistent sets of variables were reported for lakes. The data sets provided to us did not have morphometric (lake size, depth, watershed size, etc.) or hydrologic (runoff, residence times, lake type, etc.) data, except that the Ontario data set included lake size for the majority of the lakes. The Canadian investigators invested significant efforts into ensuring the quality of their data and were consulted to assist in resolving any inconsistencies that were identified as a result of our QA procedures.

Lake data were included or excluded in a specific analysis based on three types of considerations: (1) QA checks; (2) available major ion/solute chemistry data; and (3) stratification criteria. Summary statistics for pH,  $\text{SO}_4^{2-}$ , and ANC are based on nearly all lakes for which these three variables were available. Further analysis was performed on 47% of the lakes for which additional lake chemistry variables ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , DOC,  $\text{Cl}^-$ ,  $\text{Na}^+$ , and  $\text{K}^+$ ) were available to evaluate acid-base chemistry. Less than 1% of the data were omitted as a result of QA checks.

#### 2.1.1 QA Checks

Extensive QA checks were performed on the data from Canada including examining the data for potential biases. QA procedures and criteria were patterned after those established within the U.S. National Surface Water Survey projects. Single-variable range checks and visual checks for bivariate inconsistencies (e.g., low pH with high ANC) were performed. The QA screening procedure also eliminated data based on analytical techniques (1) that were not suitable (e.g., colorimetric measurements of sulfate in waters having high natural color), (2) for which change of measured cations did not balance that of measured anions, or (3) for which measured and calculated conductivities did not agree. A complete description of the QA criteria used for acceptance of data for the electroneutrality and conductivity checks are given in Appendix A. Briefly, the exclusion criteria were a function of concentration or conductivity, with the criteria being more restrictive for more concentrated lake-water samples.

#### 2.1.2 Available Major Ion/Solute Data

Most lakes have reported values for pH, ANC, or  $\text{SO}_4^{2-}$ ; however, many of the lakes are lacking values for one or more key ions, dissolved organic carbon (DOC)/color, or conductivity. Only those lakes having data for pH, ANC,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ , DOC/color, and conductivity were used in the analyses involving major solutes. These

ions provide a basis for evaluating the solutes responsible for the acid-base chemistry of lakes in southeastern Canada.

### 2.1.3 Stratification

We analyzed the entire population of sampled lakes and also stratified them into subpopulations that may be more sensitive to acidic deposition. Stratification criteria include (1) high sensitivity (low ANC of lake water) and (2) high deposition levels. These stratifications are essential to elucidate the relationships between deposition and lake chemistry and to analyze more homogeneous populations. Conclusions derived from the subpopulations only apply to similar subgroups not to the entire population of lakes.

The data screening resulted in three levels as described in the following.

#### Level 1

The Level 1 data set includes lakes for which either pH, ANC, or  $\text{SO}_4^{2-}$  values are available (Table 1) and is used to characterize lake-water chemistry of the sampled lakes. Not all lakes in Level 1 have measurements for pH, ANC, and  $\text{SO}_4^{2-}$ ; thus, the sample size in Level 1 is different for these three variables. These data have the least screening for potential analytical errors or unknown local influences (e.g., road salt and acid mine drainage).

#### Level 2

The Level 2 data set includes lakes for which pH, ANC,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and DOC values are available (Table 1). These data may still contain outliers caused by local influences (e.g., road salt, and acid mine drainage). However, the Level 2 data were not used in the analyses that follow.

#### Level 3

The Level 3 data set includes lakes for which pH, ANC,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{Cl}^-$  (sometimes estimated),  $\text{K}^+$ , DOC, and conductivity values are available (Table 1). These data were used to examine in detail the relationships between and among variables, including the charge balance and the influence of organic compounds. Lakes suspected of being influenced by road salt were excluded based on  $\text{Na}^+$  concentrations  $>87 \mu\text{eq L}^{-1}$  after sea-salt correction [Neary et al. (1990) used an exclusion concentration of  $>50 \mu\text{eq L}^{-1}$  for lakes in Ontario]. If DOC was absent for lakes in Ontario, we estimated DOC from color values by using a regression between DOC and color for lakes for which both variables were available. Missing  $\text{Cl}^-$  values were estimated from  $\text{Na}^+$  in Ontario by using a regression between  $\text{Na}^+$  and  $\text{Cl}^-$  for lakes for which both variables were available. For lakes in provinces other than Ontario, sea-salt corrections were made for  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SO}_4^{2-}$ ; for the method used to correct for sea salts (see Appendix A). This set is most like the National Surface Water Survey data used

Table 1. Number of sampled lakes in southeastern Canada by screening levels based on QA/QC checks and data completeness

	Number of Lakes						
	ON	QU	NB	NS	PE	LB	ALL
<b>First Screening<sup>a</sup></b>							
Starting	5366	2133	193	441	4	369	8506
Lakes >2000 ha	146						146
Key variables - pH, ANC, SO <sub>4</sub> <sup>2-</sup>							
All missing	2	3	0	2	0	0	7
Inconsistent pH/ANC	11	2	0	0	0	0	13
Inconsistent variables	<u>0</u>	<u>6</u>	<u>0</u>	<u>1</u>	<u>0</u>	<u>0</u>	<u>7</u>
Total Level 0 (data not used)	159	11	0	3	0	0	173
<b>Second Screening<sup>b</sup></b>							
Starting (Total Level 1)	5207	2122	193	438	4	369	8333
Key variables -- All above plus Ca <sup>2+</sup> , Mg <sup>2+</sup>							
Any one missing	2117	541	0	1	1	48	2708
ANC >2000 µeq L <sup>-1</sup>	141	2	0	0	0	0	143
pH >8	<u>77</u>	<u>16</u>	<u>2</u>	<u>0</u>	<u>0</u>	<u>0</u>	<u>95</u>
Total rejected in second screening	2335	559	2	1	1	48	2946
<b>Third Screening<sup>c</sup></b>							
Starting (Total Level 2)	2872	1563	191	437	3	321	5387
Key variables -- All above plus Na <sup>+</sup> , K <sup>+</sup> , DOC, conductivity <sup>d</sup>							
Any one missing	422	17	4	60	0	3	506
Road salt influence Na <sup>+</sup> >87 µg L <sup>-1e</sup>	111	26	2	117	3	20	279
Charge imbalance <sup>f</sup>	71	229	20	13	0	74	407
Conductivity imbalance <sup>f</sup>	<u>17</u>	<u>158</u>	<u>0</u>	<u>1</u>	<u>0</u>	<u>2</u>	<u>178</u>
Total rejected in third screening	621	430	26	191	3	99	1370
Total Level 3	2251	1133	165	246	0	222	4017

<sup>a</sup>If an observation is rejected during a first screening criteria, that observation is output to Level 0, otherwise it is output to Level 1.

<sup>b</sup>If an observation is rejected during a second screening criteria, that observation remains in Level 1, otherwise it is output to Level 2.

<sup>c</sup>If an observation is rejected during a third screening criteria, that observation remains in Level 2, otherwise it is output to Level 3.

<sup>d</sup>Missing Cl<sup>-</sup> was estimated from Na<sup>+</sup> in Ontario and not required in the other provinces.

<sup>e</sup>The value of 87 µg L<sup>-1</sup> was selected based on Na<sup>+</sup> levels and sources in Ontario; this criteria may need to be reevaluated for other provinces.

<sup>f</sup>See Appendix A.

to analyze lake-water chemistry in the United States (Baker 1990). Most lakes affected by local characteristics (e.g., road salt or acid mine drainage) were identified and excluded. Medians and interquartile ranges for pH, ANC, and  $\text{SO}_4^{2-}$  were compared between Level 1 and Level 3 data to see if QA/QC checks and stratification criteria (e.g., very large size or very high ANC) introduced a significant bias.

## 2.2 Regional Data

We adopted the regional stratification scheme developed by the Canadian scientists to build on their study of potential stratification factors (Cowell 1986, Gilbert et al. 1985, Environment Canada 1988). The basic spatial unit is the tertiary hydrologic unit defined by drainage patterns. Southeastern Canada contains 5 major river systems divided into secondary river basins and subdivided into 415 tertiary watershed units. Aggregating tertiary watersheds into larger regions was performed by Environment Canada to provide a physiographic basis for a regional framework to study the factors associated with lake-water chemistry. As a general indicator of the influence of geology on water chemistry, lakes were segregated into specific conductance classes (<25, 25 to 49, 50 to 99, and >100  $\mu\text{S cm}^{-1}$ ), and the classes were then plotted on maps. The general regional groupings suggested by these maps when combined with knowledge of geology, terrain characteristics, climate, and atmospheric deposition (including consideration of both long range and local sources) led to specification of 22 tertiary watershed aggregates across southeastern Canada (Table 2 and Fig. 1) (D. S. Jeffries, Environment Canada, personal communication, August 1989). Maps of southeastern Canada showing atmospheric deposition (Fig. 2), the potential of soil and bedrock to reduce acidity (Fig. 3), and wetlands (Fig. 4) are provided for reference.

The Belle Isle, Newfoundland (watershed aggregate 9), and Southern Ontario (aggregate 18) regions were excluded due to the predominance of limestone bedrock. Lakes in Avalon, Newfoundland (aggregate 5) were deleted due to local urban influences (D. S. Jeffries, Environment Canada, personal communication, August 1989).

For this report, we further grouped the remaining 19 aggregates into 10 regions for statistical analysis and graphical presentation (Fig. 1 and Table 2). Our ten regions consist of seven regions (1-6 and 10) in Ontario and Quebec that correspond directly to 7 of the 22 watershed aggregates defined by Environment Canada. The 12 watershed aggregates representing the Atlantic provinces were then combined into 3 contiguous regions (7, 8 and 9) based on atmospheric deposition patterns and number of Level 3 sampled lakes within each watershed aggregate. Nine of the 12 maritime watershed aggregates had fewer than 100 sampled lakes, so that combining these into larger regions reduced the possibility of sampling biases.

The regional stratification scheme, either the original Canadian aggregation, our additional aggregations, or any alternative scheme proposed for survey data lacking a sampling design, may result in aggregating lakes in regions that have distinctly different watershed

Table 2. Medians and quartiles of lake-water conductivity and wet  $\text{SO}_4^{2-}$  deposition (sea-salt corrected) for Level 3 sampled lakes by regions and aggregated tertiary watersheds (D. Jeffries, personal communication 1989) in southeastern Canada

Region	Aggregated watershed	Level 3 lakes	Conductivity ( $\mu\text{s}/\text{cm}$ )			Wet $\text{SO}_4^{2-}$ deposition ( $\text{kg}/\text{ha}$ )		
			Median	Q <sub>1</sub>	Q <sub>3</sub>	Median	Q <sub>1</sub>	Q <sub>3</sub>
1-West Ontario	21	262	32	27	42	8	9	
2-North Ontario	20	397	76	46	118	15	17	
3-Southeast Ontario	17	968	31	26	36	26	27	
4-Ottawa Valley	16	83	34	25	49	22	24	
5-Southeast Quebec	15	513	32	27	40	23	23	23
6-Laurentide	14	442	21	18	25	21	20	22
7-North Quebec	13	274	19	14	25	14	16	
	22	<u>42</u>	<u>16</u>	<u>12</u>	<u>21</u>	<u>16</u>	<u>13</u>	<u>20</u>
		316	19	14	25	14	13	16
8-South Atlantic	1	241	26	22	31	15	15	15
	2	44	34	27	44	14	14	14
	3	98	30	22	45	14	13	15
	4	28	26	18	30	13	13	13
	12	<u>37</u>	<u>99</u>	<u>41</u>	<u>118</u>	<u>26</u>	<u>22</u>	<u>29</u>
		448	28	23	29	14	14	15
9-North Atlantic	6	48	26	21	32	8	9	
	7	34	18	15	21	11	9	12
	8	60	24	19	28	10	10	11
	10	102	12	10	14	7	6	9
	11	<u>15</u>	<u>13</u>	<u>12</u>	<u>23</u>	<u>5</u>	<u>5</u>	<u>6</u>
		259	18	12	24	9	7	10
10-Sudbury/Noranda	19	311	36	32	41	21	18	24

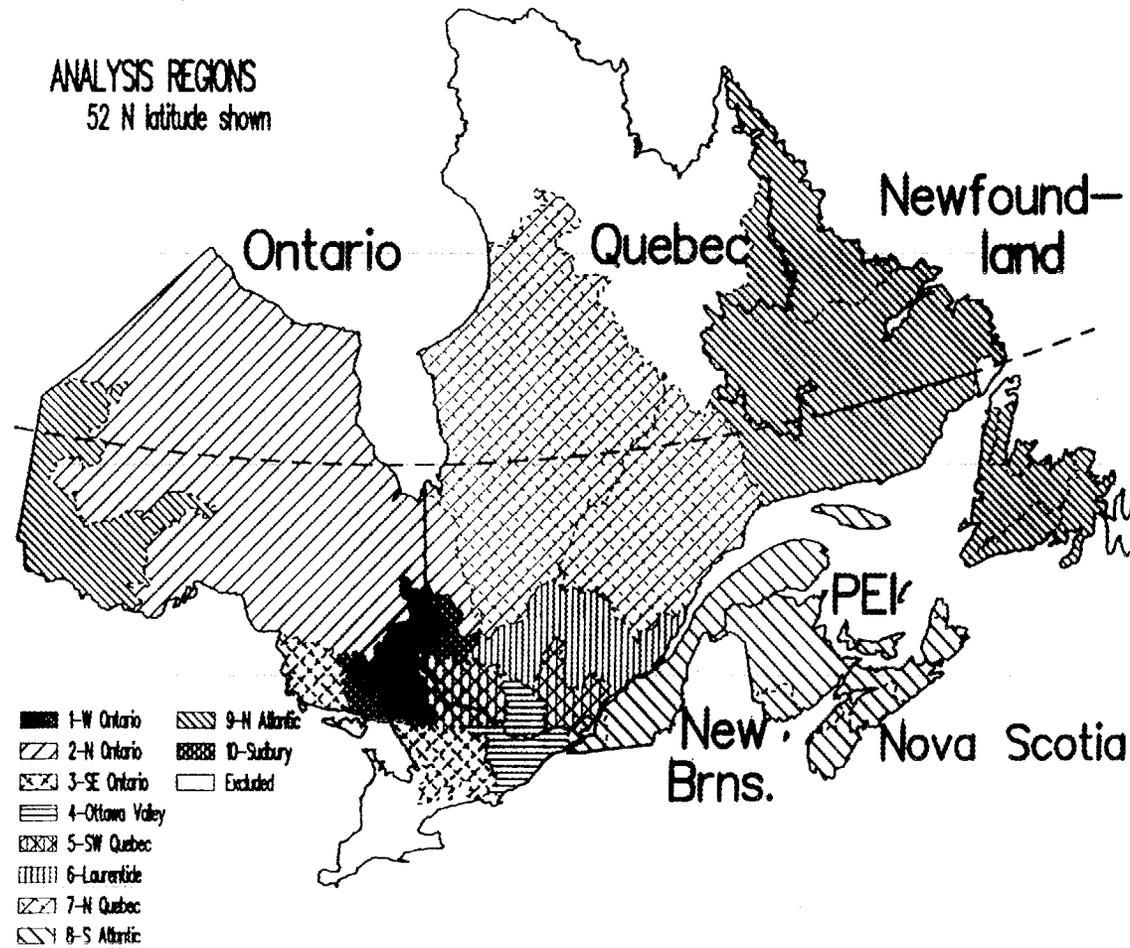


Fig. 1. Map of southeastern Canada showing regions defined and used in this report. Provincial boundaries, boundaries of aggregated watersheds, and 52°N latitude are also shown.

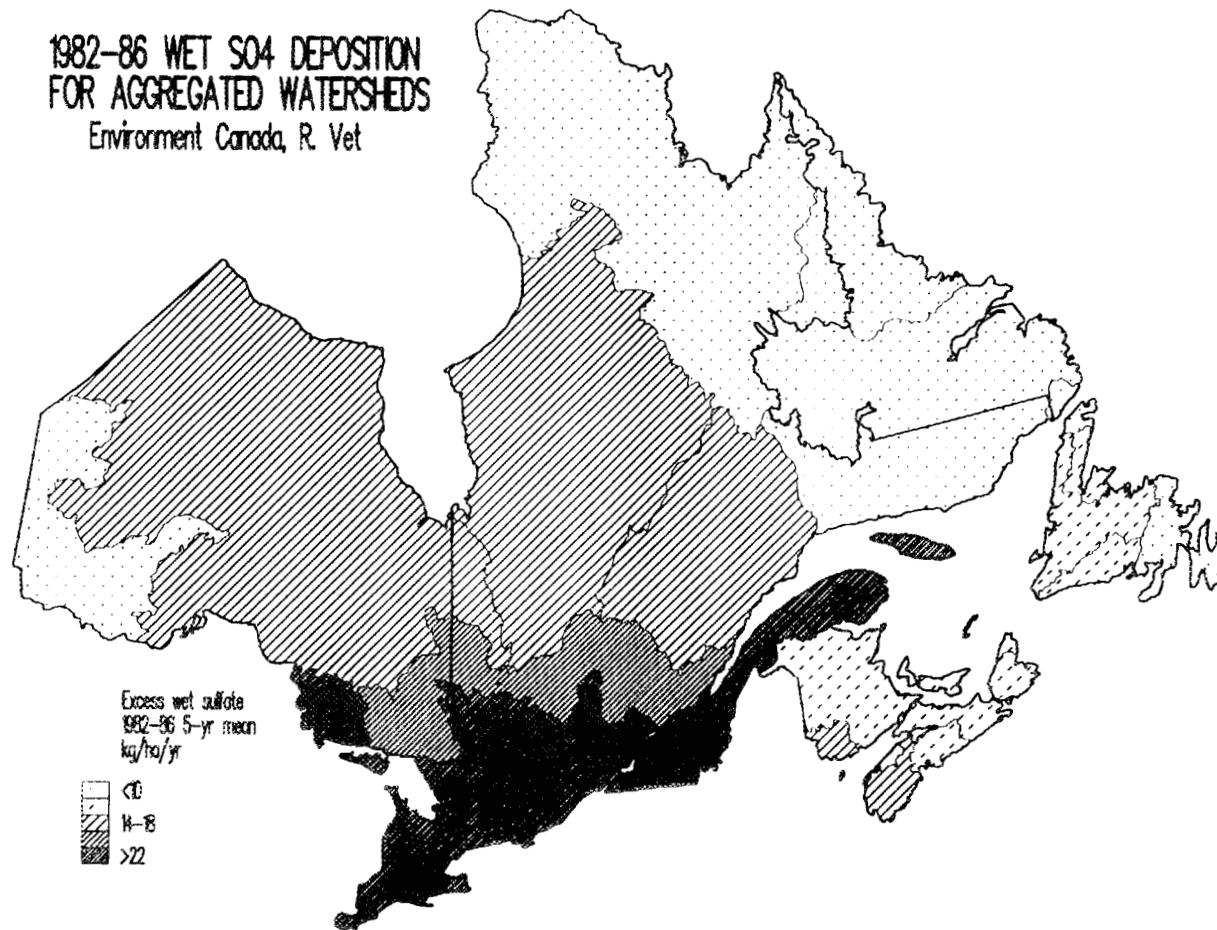


Fig. 2. Map of 1982-1986 mean annual wet SO<sub>4</sub><sup>2-</sup> deposition (sea salt corrected) in southeastern Canada. Data from R. Vet, Environment Canada, personal communication, October 1989.

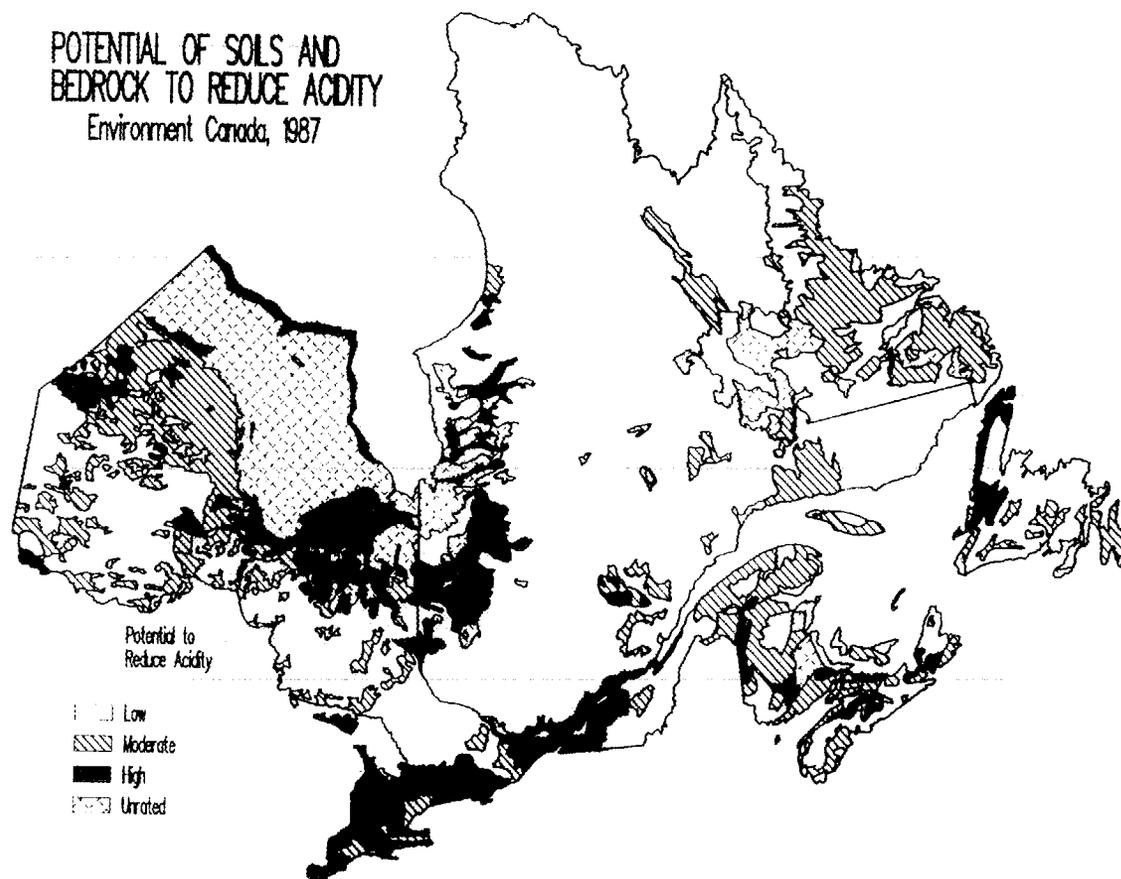


Fig. 3. Map of potential of soils and bedrock to reduce acidity of atmospheric deposition in Canada. Areas are defined as: low—areas primarily composed of noncarbonate bedrock and coarse textured shallow soils; moderate—areas primarily consisting of noncarbonate bedrock and/or shallow to deep soils; high—areas primarily consisting of carbonate bedrock and/or deep, fine textured soils; and unrated—dominated by peatlands. Source: Environment Canada, 1988.

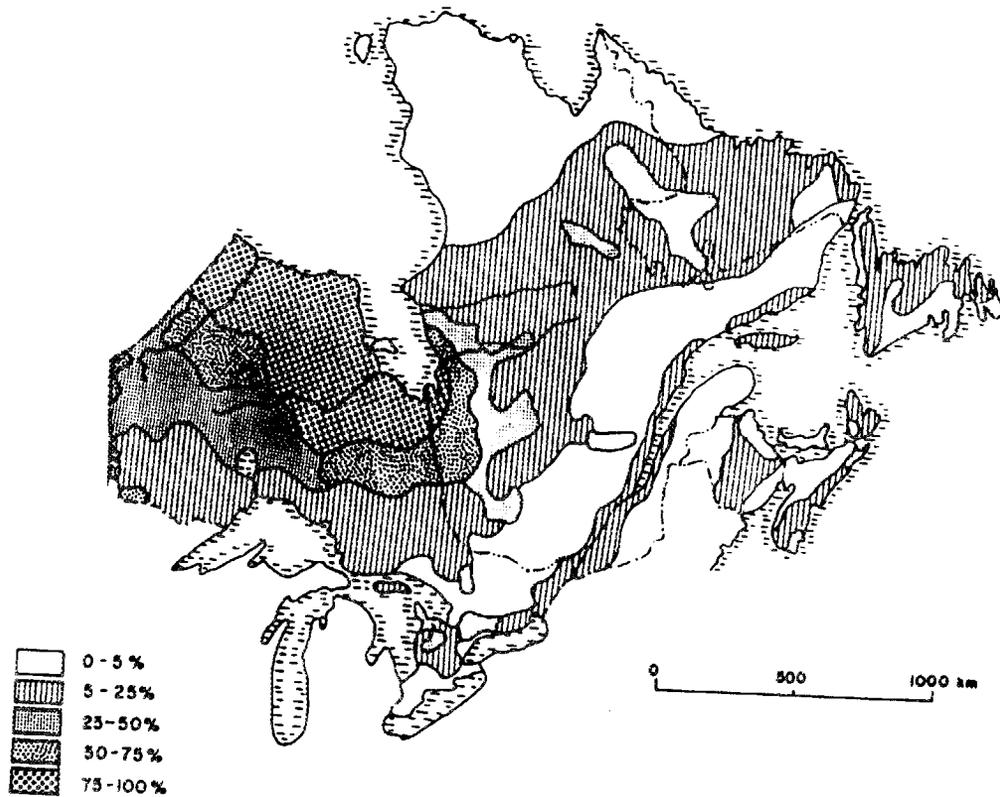


Fig. 4. Distribution of wetlands in Canada, expressed as percentage of total land surface. Wetlands are defined as areas where wet soils are prevalent, having a watertable near or above the mineral soil. Source: Zoltai and Pollett, 1983.

attributes (e.g. atmospheric deposition sources or geology types). Although the stratification scheme allows us to examine dominant regional patterns across southeastern Canada, variance of regional statistics may be increased as a result of local sources of variation. The contribution from factors within a region to the regional pattern must be interpreted with caution. For example, the Sudbury-Noranda region is larger than the area that has been estimated as under the local influence of the smelters (Neary et al. 1990), and therefore that region includes both lakes subjected to high local  $\text{SO}_4^{2-}$  deposition and lakes receiving lower  $\text{SO}_4^{2-}$  deposition loadings from long-range transport. Watershed aggregate 12, St. Lawrence South Shore, has only 37 Level 3 lakes and presented us with the dilemma of choosing whether to retain the area as a separate region or to combine it with other appropriate watershed aggregates. This area has bedrock differing from adjacent watershed aggregates and relatively high atmospheric deposition; we combined it with the South Atlantic Region (Region 8).

Regional lake population data were obtained from Environment Canada (D. S. Jeffries, personal communication, September 1989). Numbers and areas of surface waters in southeastern Canada, including location, surface area, and shoreline length for lakes with surface area  $>0.18$  ha, were determined from remote imagery (Hélie 1989). The inventory data covered an area south of  $52^\circ\text{N}$  and east of  $90^\circ\text{W}$ , with some incomplete coverage of southern Ontario and Nova Scotia. Population numbers and lake areas were summarized for tertiary watershed units based on lakes  $>1$  ha.

Wet  $\text{SO}_4^{2-}$  deposition data, consistent with deposition data for the United States being used for the 1990 NAPAP SOS/T, were obtained from Environment Canada for southeastern Canada south of  $52^\circ\text{N}$  (R. Vet, Environment Canada, personal communication, October 1989). These deposition data were expressed as average excess  $\text{SO}_4^{2-}$  (i.e., sea-salt corrected) for the 1982-1986 period for  $0.5^\circ$  grid cells for southeastern Canada (Fig. 2). Grid files were generated for each year using kriging to interpolate from monitoring sites; grid values from each of the years were averaged to produce the 5-year means.

### 3. RESULTS AND DISCUSSION

#### 3.1 Data Bases for Lake Chemistry Data

Over 376,000 lakes are  $>1$  ha in size in southeastern Canada, with a total surface area of over 18 million ha (Table 3, Hélie 1989). Southeastern Canada, as defined by south of  $52^\circ\text{N}$  and east of  $90^\circ\text{W}$ , represents 16% of the area of Canada. Numbers of lakes and lake areas are presented in Table 3 for those five regions for which the inventory was complete. Five regions for which the coverage was incomplete, because the region either extended north of  $52^\circ\text{N}$  or west of  $90^\circ\text{W}$ , were classified into the "other" category. Most (94%) of the

Table 3. Numbers of lakes and total lake surface areas by size classes for regions in southeastern Canada and estimated percentage of lakes >1 ha sampled for lake-water chemistry. Regions with incomplete data are grouped in the "other" class. Data are from analysis of remote imagery (Hélie 1989) covering eastern Canada south of 52°N and east of 90°W.

Region	1-5 ha		5-50 ha		50-100 ha		100-500 ha		>500 ha		Total		Estimated percentage sampled
	N	ha	N	ha	N	ha	N	ha	N	ha	N	ha	
3	15,235	35,135	7,309	101,080	518	36,292	460	89,928	127	267,266	23,649	529,701	9.0
4	4,560	10,573	2,515	36,618	200	14,000	235	46,085	89	142,838	7,599	250,114	3.3
5	10,539	26,072	7,906	116,522	662	45,248	498	103,264	110	220,351	19,715	511,458	3.8
6	15,737	39,425	12,389	178,640	914	63,244	656	128,670	145	580,984	29,841	990,964	2.5
10	11,592	27,130	6,482	95,151	554	38,787	439	89,670	133	448,090	19,200	698,828	3.7
Other <sup>1</sup>	163,835	389,044	96,897	1,398,651	7,909	548,628	6,242	1,245,271	1,352	12,129,438	276,235	15,711,032	
All	221,498	527,378	133,498	1,926,663	10,757	746,200	8,530	1,702,888	1,956	13,788,968	376,239	18,692,096	

<sup>1</sup>Other regions not completely covered by the inventory of lakes, which covered eastern Canada south of 52°N and east of 90°W.

lakes are <50 ha in surface area, whereas the lakes >500 ha represent 74% of total lake surface area.

Lake-water chemistry data for 5366 Ontario lakes were screened for QA/QC and analyzed. The screening process identified 5207 (97%) lakes having either pH, ANC, or  $\text{SO}_4^{2-}$  data (Level 1), whereas 2251 of these lakes (43%) had additional variables (Level 3) to allow more-detailed examination of acid-base chemistry on a regional scale (Table 1).

Data were obtained for 3140 lakes in the remainder of southeastern Canada (Quebec, Newfoundland, Nova Scotia, and New Brunswick). The screening process identified 3126 (99.6%) of these qualifying as Level 1 and 1766 (56%) of these as Level 3 (Table 1). Sea-salt corrections were made for  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SO}_4^{2-}$  using equations provided with the data (D. S. Jeffries, Environment Canada, personal communication, September 1989).

The 8333 Level 1 lakes are concentrated in the southwestern Quebec and southeastern Ontario regions (Fig. 5) with the 4017 Level 3 lakes showing a geographic distribution broadly similar to that of the Level 1 lakes (Fig. 6). Even though the sampling density in Nova Scotia, insular Newfoundland, and Labrador is less than that in southern Ontario and Quebec, the spatial coverage in these southeastern Canada provinces is thorough.

A comparison of Level 1 and Level 3 data (Table 4) shows that both the 25th percentile ANC values and the median ANC values are similar, except for the Ottawa Valley (Region 4) and Sudbury/Noranda (Region 10). For these two levels, the median pH and  $\text{SO}_4^{2-}$  values are also similar for all regions in southeastern Canada (Tables 5 and 6).

Many of the lakes in southeastern Canada do not have DOC or color data. To include the largest possible number of lakes that had a complete set of variables and to conduct QA checks, DOC was estimated from color values for 808 lakes in Ontario. A linear regression was developed for those Level 3 lakes ( $n = 1567$ ) for which we had DOC and apparent color data ( $\text{DOC} = 2.2115 + 0.0927 \times \text{Apparent Color}$ ) that had an  $R^2 = 0.6$ . Means and standard deviations of DOC calculated from color were in close agreement with those for lakes with measured DOC. Using a similar approach, Neary et al. (1990) calculated separate regression equations for latitudinal bands to account for an observed trend of increasing residuals with latitude.

$\text{Cl}^-$  was estimated for 2491 lakes in Ontario using a linear regression for  $\text{Na}^+$  and  $\text{Cl}^-$  from 258 Level 3 lakes ( $\text{Cl}^- = -1.8962 + 0.32 \times \text{Na}^+$ ); the  $R^2$  for this regression was 0.2. Lakes for which estimated  $\text{Cl}^-$  data were available had significantly higher pH, ANC, and  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  than those for which we had measured  $\text{Cl}^-$  data. Because the range of  $\text{Na}^+$  in calibration data was similar to estimated  $\text{Na}^+$  and because  $\text{Cl}^-$  is typically present at low concentrations and is a minor anion, we decided to use the estimated  $\text{Cl}^-$  values for charge balance checks and conductivity balance checks.  $\text{Cl}^-$  is not used directly in any of the analyses.

The annual wet excess  $\text{SO}_4^{2-}$  deposition (i.e., sea-salt corrected) was highest in the southeastern area of Ontario and southwestern area of Quebec and decreased to the north, east, and west (Table 2 and

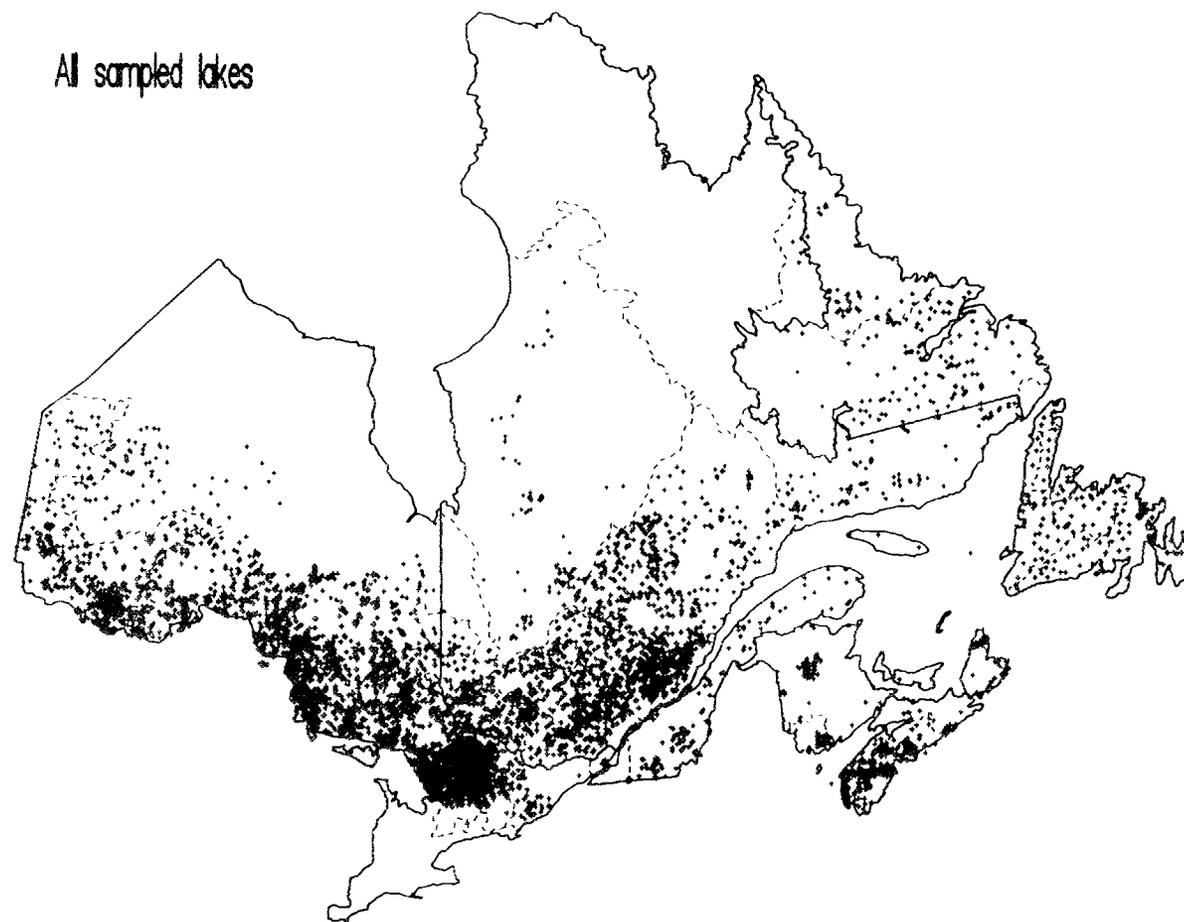


Fig. 5. Map of 8333 Level 1 sampled lakes in southeastern Canada.

Level 3 lakes

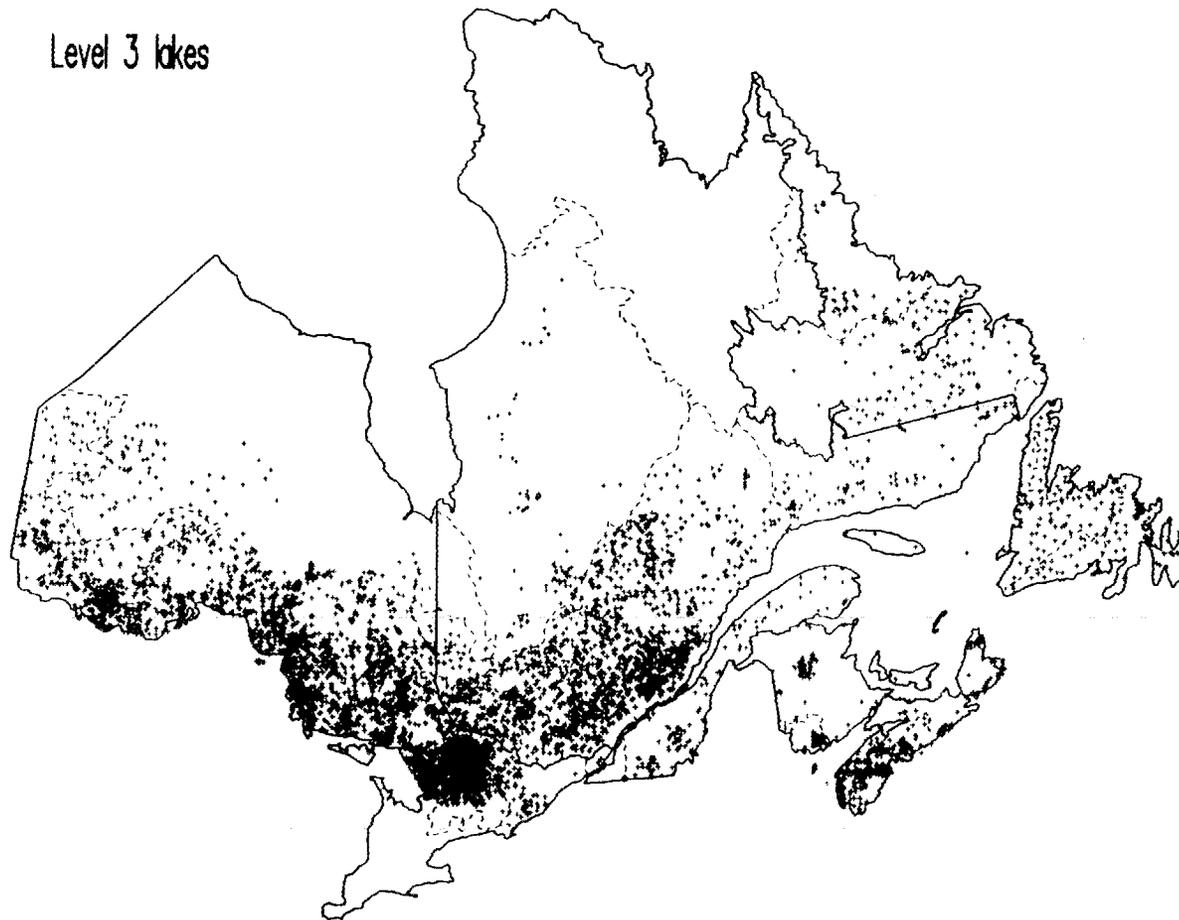


Fig. 6. Map of 4017 Level 3 sampled lakes in southeastern Canada.

Table 4. Acid neutralizing capacity (ANC) of sampled lakes in southeastern Canada by region for levels 1 and 3 screening subsets including number (n), 25th percentile (Q<sub>1</sub>), median, and 75th percentile (Q<sub>3</sub>)

Region	Level	ANC ( $\mu\text{eq/L}$ )			
		n	Q <sub>1</sub>	Median	Q <sub>3</sub>
1-West Ontario	1	568	108	172	295
	3	265	111	173	275
2-North Ontario	1	1382	287	640	1326
	3	397	234	592	1093
3-Southeast Ontario	1	2086	30	62	124
	3	974	23	49	97
4-Ottawa Valley	1	236	68	203	903
	3	83	60	115	308
5-Southwest Quebec	1	747	38	78	153
	3	513	40	79	144
6-Laurentide	1	734	26	44	76
	3	442	28	45	74
7-North Quebec	1	538	27	60	124
	3	316	30	71	136
8-South Atlantic	1	764	8	32	123
	3	448	6	24	73
9-North Atlantic	1	442	24	42	76
	3	259	22	40	74
10-Sudbury/Noranda	1	678	1	33	126
	3	312	-9	14	49

Table 5. pH of sampled lakes in southeastern Canada by region for Levels 1 and 3 screening subsets including number (n), 25th percentile (Q<sub>1</sub>), median, and 75th percentile (Q<sub>3</sub>)

Region	Level	pH			
		n	Q <sub>1</sub>	Median	Q <sub>3</sub>
1-West Ontario	1	569	6.60	6.87	7.20
	3	265	6.70	6.91	7.18
2-North Ontario	1	1395	7.02	7.44	7.85
	3	397	7.04	7.40	7.73
3-Southeast Ontario	1	2121	5.89	6.29	6.73
	3	974	5.75	6.11	6.47
4-Ottawa Valley	1	253	6.20	6.90	7.97
	3	83	6.00	6.50	6.97
5-Southwest Quebec	1	752	5.97	6.31	6.70
	3	513	6.00	6.34	6.65
6-Laurentide	1	744	5.60	6.00	6.30
	3	442	5.70	6.00	6.20
7-North Quebec	1	540	5.90	6.30	6.60
	3	316	5.90	6.30	6.67
8-South Atlantic	1	765	5.25	5.91	6.61
	3	448	5.20	5.80	6.40
9-North Atlantic	1	426	5.83	6.20	6.50
	3	259	5.90	6.20	6.60
10-Sudbury/Noranda	1	706	5.30	6.01	6.67
	3	312	4.98	5.70	6.31

Table 6. Sulfate ( $\text{SO}_4^{2-}$ ) of sampled lakes in southeastern Canada by region for Levels 1 and 3 screening subsets including number (n), 25th percentile ( $Q_1$ ), median, and 75th percentile ( $Q_3$ )

Region	Level	$\text{SO}_4^{2-}$ ( $\mu\text{eq L}^{-1}$ )			
		N	$Q_1$	Median	$Q_3$
1-West Ontario	1	340	52	67	81
	3	265	52	69	81
2-North Ontario	1	785	58	91	119
	3	397	59	92	119
3-Southeast Ontario	1	1371	122	150	169
	3	974	117	150	168
4-Ottawa Valley	1	170	111	132	165
	3	83	103	123	162
5-Southwest Quebec	1	703	123	144	165
	3	513	123	144	165
6-Laurentide	1	731	60	81	102
	3	442	60	81	102
7-North Quebec	1	522	40	51	62
	3	316	40	51	62
8-South Atlantic	1	725	39	53	72
	3	448	37	50	68
9-North Atlantic	1	441	20	30	40
	3	259	21	33	40
10-Sudbury/Noranda	1	470	173	208	239
	3	312	182	210	237

Fig. 2).  $\text{SO}_4^{2-}$  deposition was estimated for individual lakes ( $n = 7857$ ) by identifying the closest grid point and assigning that value to the lake. This method of estimating deposition for individual lakes differed from the kriging method used for estimating deposition at individual lakes in the United States as part of the Eastern Lake Survey (Linthurst et al. 1986). Lakes in the areas lacking deposition data were excluded from analyses involving deposition (Sect. 2.3).

### 3.2 Characteristics of Lake-Water Chemistry for Sampled Lakes

Our analysis is conducted only for the sampled lakes with low ANC and <2000 ha, not the entire population of lakes in southeastern Canada. Because of the potential biases in the selection of Canadian lakes for sample collection, we do not know if the sampled population is representative of the entire population, and we cannot quantify the confidence intervals for estimates of the entire population of lakes. For example, small lakes, which may have a comparatively lower pH and ANC, are underrepresented (D. S. Jeffries, Environment Canada, personal communication, November 29, 1989).

Of the 8192 sampled lakes and with ANC, 4613 (56%) have an ANC  $\leq 100 \mu\text{eq L}^{-1}$  (Table 7). Sampled lakes in this strata are located throughout southeastern Canada and are located with highest density in southeast Ontario (Region 3), South Atlantic (Region 8), and southern Quebec (Regions 4-6) (Table 7 and Fig. 7). Of the Level 1 sampled lakes, 38% (3041 out of 8192) have an ANC  $\leq 50 \mu\text{eq L}^{-1}$  (Table 7). Sampled lakes with ANC  $\leq 50 \mu\text{eq L}^{-1}$  (Fig. 8) are located throughout southeastern Canada, primarily in southeastern Ontario (Region 3) and in the South Atlantic (Region 8). In part, the high numbers of low-ANC lakes in southeastern Ontario (Region 3) results from the high sampling densities; 9.0% of the lakes in southeastern Ontario were sampled, compared with 2-4% for Regions 4, 5, 6, and 10 (Table 3).

Of the Level 1 sampled lakes in southeastern Canada, 381 lakes (5% of the sampled lakes) are acidic (ANC  $\leq 0 \mu\text{eq L}^{-1}$ ) (Table 7, Fig. 9). The Sudbury/Noranda region has the greatest percentage of sampled acidic lakes (24%), and it has 43% of the sampled acidic lakes in all of southeastern Canada. The Sudbury/Noranda region also has a relatively high sampling density—an estimated 3.7% of the lakes > 1 ha in the region were sampled (Table 3). In the South Atlantic region, 90 of the 764 sampled lakes (12%) are acidic, and in southeastern Ontario, 86 (4%) of the sampled lakes are acidic. Most of the acidic lakes in the South Atlantic region occur in Nova Scotia; of the 438 sampled lakes in Nova Scotia 77 (17.5%) are acidic. No acidic lakes were sampled in the West Ontario region, and <1% of the lakes sampled in North Ontario were acidic (Table 7).

The Level 1 data base contains 20 acidic lakes in Quebec (Ottawa Valley, Southwest Quebec, Laurentide, and North Quebec). This number is probably an underestimate because the laboratories of the Ministry of the Environment, Quebec, do not report negative ANC values (J. Dupont, Ministry of the Environment, Sainte-Foy, Quebec, personal communication, February 1990). Instead, the laboratories report an ANC detection limit of  $2 \mu\text{eq L}^{-1}$ ; in the data base obtained from

Table 7. Number of level 1 sampled lakes by ANC class and region in southeastern Canada.

Region	Number of Lakes	Number of sampled lakes by ANC class					Percentage of lakes ANC $\leq 0$	Percentage of 381 lakes ANC $\leq 0$
		$\leq 400$	$\leq 200$	$\leq 100$	$\leq 50$	$\leq 0^a$		
1-West Ontario	568	467	322	121	36	0	0	0
2-North Ontario	1382	471	226	113	54	8	<1	2
3-Southeast Ontario	2086	1928	1768	1439	864	86	4	23
4-Ottawa Valley	236	145	118	83	43	1	<1	<1
5-Southwest Quebec	749	718	616	449	254	4	<1	1
6-Laurentide	743	736	715	627	445	8	1	2
7-North Quebec	538	516	465	373	239	7	1	2
8-South Atlantic	764	660	623	552	450	90	12	24
9-North Atlantic	442	434	419	373	254	14	3	4
10-Sudbury/Noranda	684	632	568	483	402	163	24	43
All	8192	6707	5840	4613	3041	381	5	100

<sup>a</sup>Seventeen lakes had a recorded ANC value of  $2 \mu\text{eq L}^{-1}$ , the detection limit of techniques used in Quebec. These lakes are located in Regions 5 (2 lakes), 6 (9 lakes), and 10 (6 lakes). If the ANC of these lakes is assumed to be  $\leq 0$  then the number of acidic lakes would be increased to 6 (1%), 17 (2%), 169 (25%), respectively.

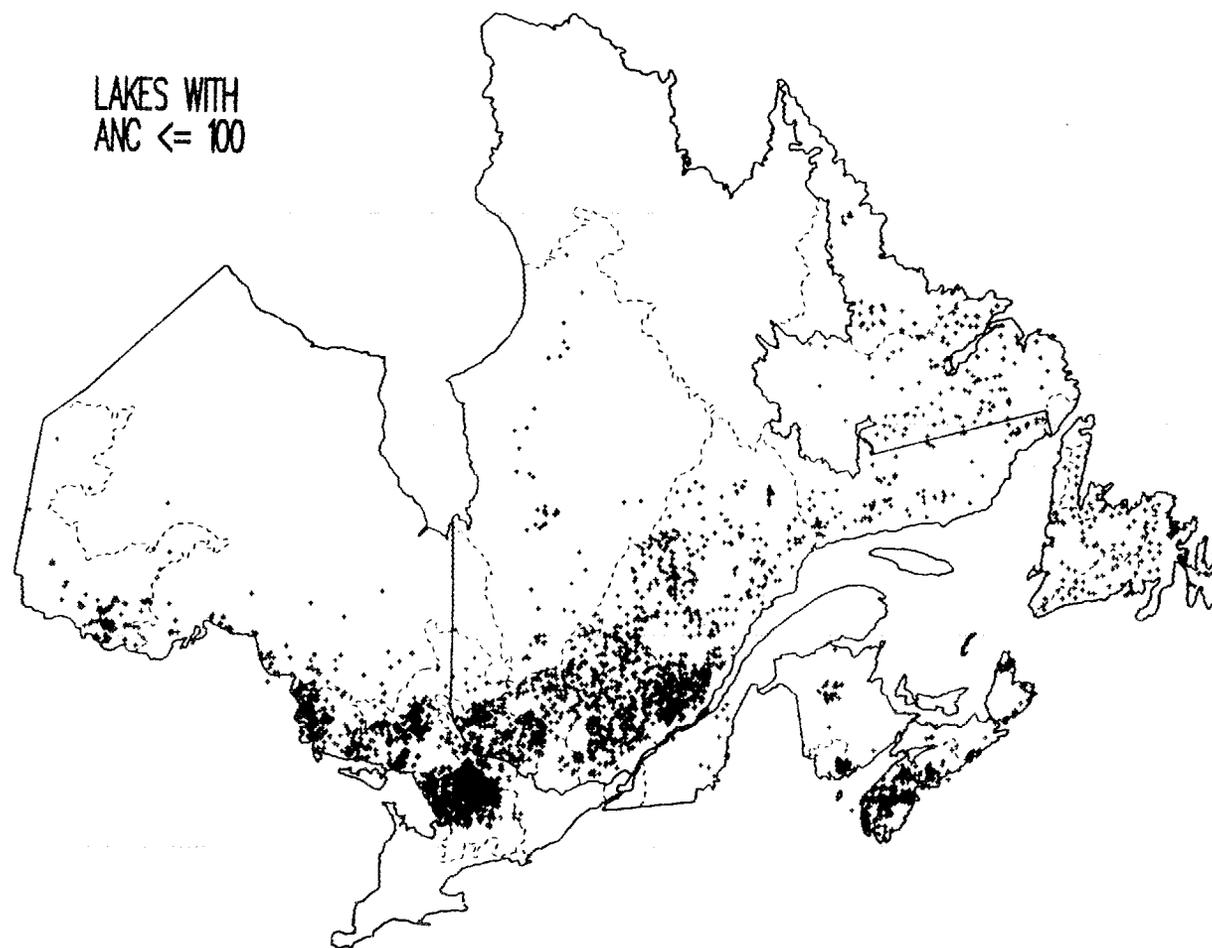


Fig. 7. Map of 4611 Level 1 sampled lakes in southeastern Canada having ANC  $\leq 100 \mu\text{eq L}^{-1}$ .

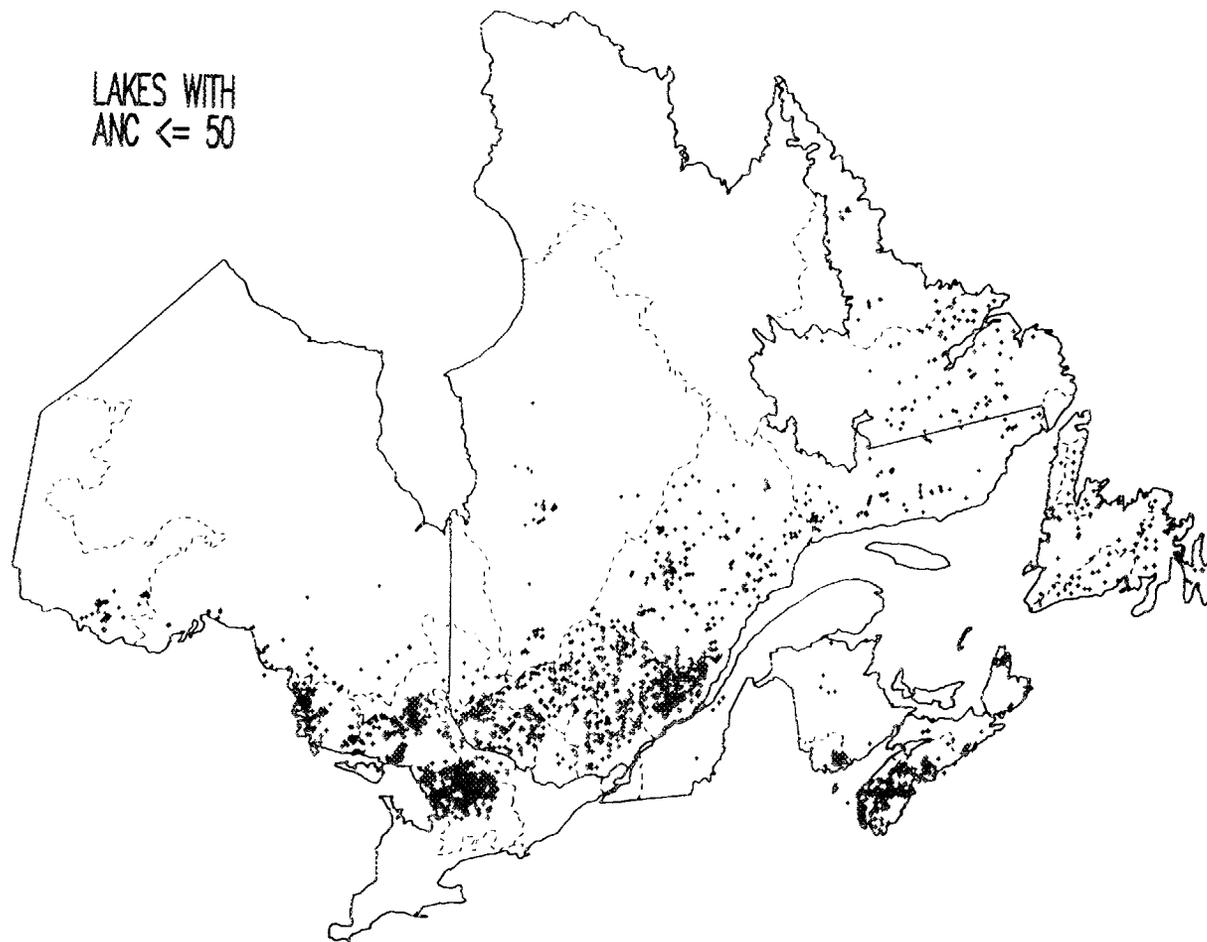


Fig. 8. Map of 3035 Level 1 sampled lakes in southeastern Canada having ANC  $\leq 50 \mu\text{eq L}^{-1}$ .

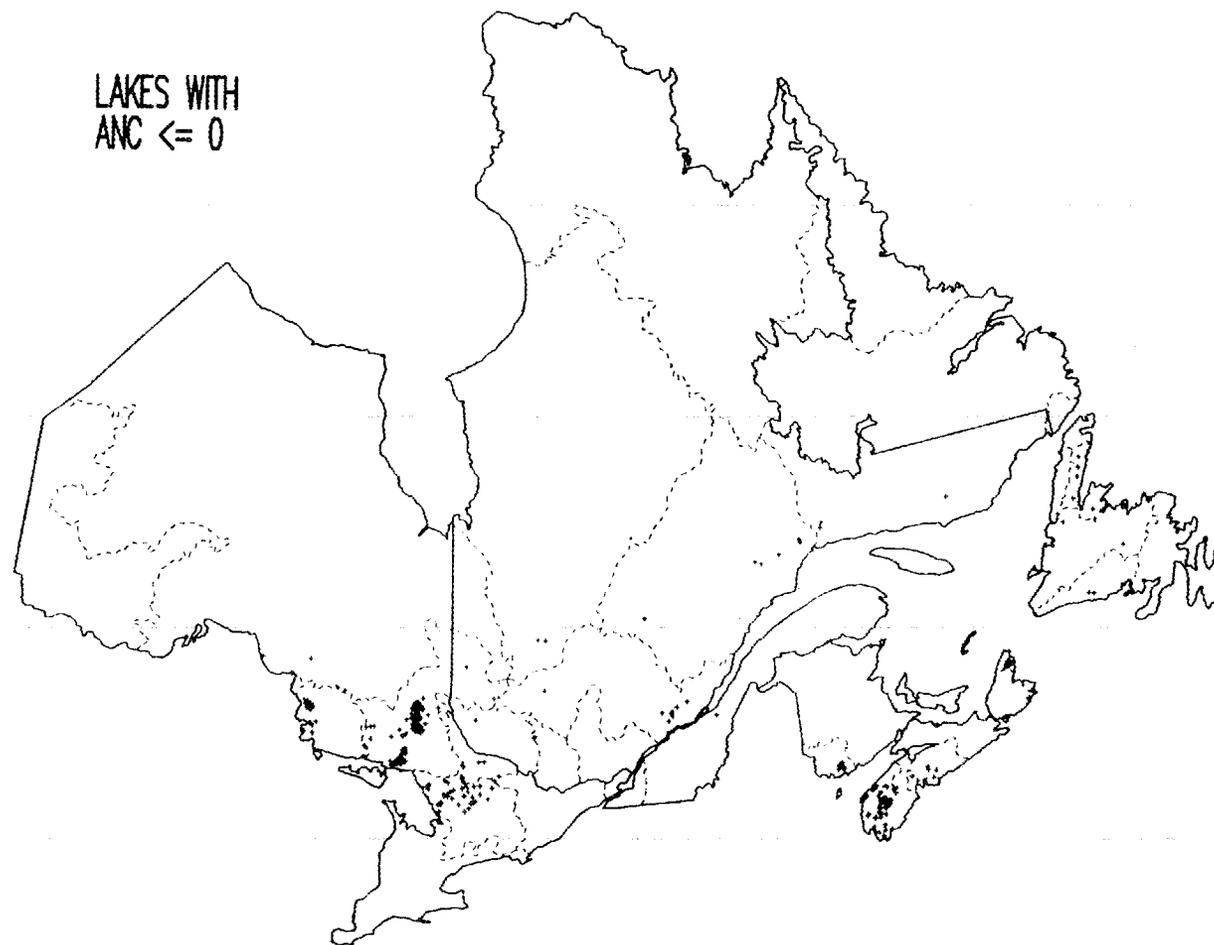


Fig. 9. Map of 381 Level 1 sampled lakes in southeastern Canada with  $ANC \leq 0 \mu\text{eq L}^{-1}$ .

Environment Canada, this value is simply reported as  $2 \mu\text{eq L}^{-1}$ , not  $\leq 2 \mu\text{eq L}^{-1}$ .

It is important to note that episodic ANC depressions are associated with snowmelt events in southeastern Canada (Jeffries in press). Because nearly all of the lakes were sampled either under ice cover prior to snow melt or during the summer months, the influence of snowmelt ANC depressions are not represented in the data presented here.

The primary variable used in this report to assess the acid-base characteristics of lakes in Canada is ANC. As described in the following, pH measurements made in southeastern Canada are affected to widely varying degrees by supersaturation with respect to carbon dioxide ( $\text{CO}_2$ ) and also by organic acids. For example, some of the lakes were sampled during periods of ice cover and are likely to have extremely high partial pressures of  $\text{CO}_2$  (Kratz et al. 1987, D. S. Jeffries, Environment Canada, personal communication, February 1990), which causes pH to be lower by as much as 1 pH unit for every factor of 10 in  $\text{CO}_2$  supersaturation (Turner et al. 1990). ANC, which is conservative with respect to  $\text{CO}_2$  is therefore the preferred variable for characterizing the acid-base status of systems possibly affected by acidic deposition.

The number of Level 1 sampled lakes with  $\text{pH} \leq 6$  is 2557 (31% of the 8296 Level 1 lakes for which pH data were available, Table 8). Sampled lakes having a pH of  $\leq 6.0$  are primarily located in Southeast Ontario (Region 3), Laurentide (Region 6), South Atlantic (Region 8), and Sudbury/Noranda (Region 10). The number of Level 1 sampled lakes having a pH of  $\leq 5.5$  is 1080 (13% of the total Level 1 lakes); these lakes are primarily located in the South Atlantic (Region 8), Southeast Ontario (Region 3), and Sudbury/Noranda (Region 10). The number of Level 1 sampled lakes having a pH of  $\leq 5.0$  is 393, or 5%. Most of the  $\text{pH} \leq 5.0$  lakes are located in the Sudbury/ Noranda and the South Atlantic regions (Table 8).

Except for the South Atlantic (median pH 5.80) and Sudbury/ Noranda regions (median pH 5.7), median pH values are circumneutral across southeastern Canada (range for other regions: 6.0 to 7.4, Table 5). Median ANC for the Level 3 lakes is lowest in the Sudbury/Noranda, southeastern Ontario, and South Atlantic regions. Median  $\text{SO}_4^{2-}$  concentrations are highest in Sudbury/Noranda, southeastern Ontario, Ottawa Valley, and southwest Quebec regions (Table 6). The lowest median  $\text{SO}_4^{2-}$  concentrations are found in North Atlantic (Region 9), an area that receives low levels of atmospheric deposition. For the Level 3 lakes, the median sum of base cations ( $\text{SBC} = \text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+$ ) and sum of calcium and magnesium [ $\text{Ca}^{2+} + \text{Mg}^{2+}$ ] are highest in North Ontario, West Ontario, and the Ottawa Valley and lowest in the North Atlantic and South Atlantic regions (Table 9). Median DOC concentrations are  $>300 \mu\text{mol L}^{-1}$  for all regions, except the Sudbury/Noranda, and are highest in North Ontario, West Ontario, Laurentide, and North Atlantic (Table 10). Wetlands, a source of DOC to surface waters, cover between 25 and 100% of the landscape in North Ontario and  $>5-50\%$  in West Ontario (Fig. 4) (Zoltai and Pollett 1983).

Examination of lake-water chemistry for lakes having ANC levels of  $\leq 400 \mu\text{eq L}^{-1}$  reveals a decrease in median ANC values from

Table 8. Number of Level 1 sampled lakes by pH class and region in southeastern Canada

Region	Number of Lakes	Number of sampled lakes by pH Class			Percentage of lakes pH $\leq 5.0$	Percentage of 393 lakes pH $\leq 5.0$
		$\leq 6.0$	$\leq 5.5$	$\leq 5.0$		
1-West Ontario	569	21	6	0	0	0
2-North Ontario	1395	56	25	12	1	3
3-Southeast Ontario	2121	652	227	47	2	12
4-Ottawa Valley	253	48	10	3	1	1
5-Southwest Quebec	752	214	70	16	2	4
6-Laurentide	744	436	140	33	4	8
7-North Quebec	540	188	54	13	2	3
8-South Atlantic	765	416	274	121	16	31
9-North Atlantic	426	173	55	12	3	3
10-Sudbury/Noranda	706	353	219	136	19	35
All	8271	2557	1080	393	5	100

Table 9. Sum of base cations (SBC) and sum of calcium and magnesium [ $\text{Ca}^{2+} + \text{Mg}^{2+}$ ] for sampled lakes in southeastern Canada by region for Level 3 screened subsets including number (n), 25th percentile ( $Q_1$ ), median, and 75th percentile ( $Q_3$ )

Region	$(\mu\text{eq L}^{-1})\text{SBC}$				$(\mu\text{eq L}^{-1})\text{Ca}^{2+} + \text{Mg}^{2+}$			
	n	$Q_1$	Median	$Q_3$	n	$Q_1$	Median	$Q_3$
1-West Ontario	265	237	320	419	265	182	273	332
2-North Ontario	397	433	772	1204	397	385	730	1137
3-Southeast Ontario	974	193	234	289	974	159	192	239
4-Ottawa Valley	83	202	289	468	83	160	242	413
5-Southwest Quebec	513	209	266	332	513	169	220	273
6-Laurentide	442	142	171	209	442	113	137	166
7-North Quebec	316	116	160	232	316	89	130	193
8-South Atlantic	448	111	141	203	448	60	81	140
9-North Atlantic	259	78	113	157	259	55	86	112
10-Sudbury/Noranda	312	197	245	302	312	156	200	250

Table 10. Dissolved organic carbon (DOC) and chloride ( $\text{Cl}^-$ ) for sampled lakes in southeastern Canada by region for Level 3 screened subsets including number (n), 25th percentile ( $Q_1$ ), median, and 75th percentile ( $Q_3$ )

Region	DOC ( $\mu\text{mol L}^{-1}$ )				Cl <sup>-</sup> ( $\mu\text{eq L}^{-1}$ )			
	n	$Q_1$	Median	$Q_3$	n	$Q_1$	Median	$Q_3$
1-West Ontario	265	292	400	566	265	6.39	9.94	13.42
2-North Ontario	397	425	650	925	397	5.76	7.85	10.14
3-Southeast Ontario	974	275	350	466	974	5.06	7.15	9.24
4-Ottawa Valley	69	296	416	533	83	11.28	14.10	16.93
5-Southwest Quebec	502	279	367	483	513	8.54	11.28	14.13
6-Laurentide	394	300	427	566	442	11.28	14.10	16.93
7-North Quebec	233	354	483	604	316	5.64	8.46	14.10
8-South Atlantic	340	262	367	564	448	56.77	101.53	141.05
9-North Atlantic	123	292	399	579	259	14.10	50.78	84.63
10-Sudbury/Noranda	312	192	267	400	312	6.46	8.54	11.57

west-to-east, except for southeastern Ontario (Fig. 10). Similarly, the median  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  decreases from west-to-east. Median  $\text{SO}_4^{2-}$  concentrations increase from West Ontario to the central portion of southeastern Canada (Sudbury/Noranda, southeast Ontario, Ottawa Valley, and southwest Quebec) and decrease farther eastward to the North Atlantic region (Fig. 10). This pattern of lake-water  $\text{SO}_4^{2-}$  reflects the geographic distribution of  $\text{SO}_4^{2-}$  deposition in the central portion of southeastern Canada (see Sect. 2.3). In addition, a bedrock source of sulfur in the Ottawa Valley region contributes to lake-water  $\text{SO}_4^{2-}$ , although the lakes influenced have a higher ANC and, thus, are not sensitive to the effects of atmospheric deposition (J. Dupont, Ministry of the Environment, Sainte-Foy, Quebec, personal communication, February 1990).

Organic anion concentrations were estimated from pH and DOC based on Oliver's model (Oliver et al. 1983), which uses an acid functional group content of  $10 \mu\text{eq} (\text{mg DOC})^{-1}$ . Recent compilations of detailed characterization of the acid function group content suggest that the range is from 5 to  $15 \mu\text{eq} (\text{mg DOC})^{-1}$  (Cook et al. 1987, Urban et al. 1989); estimates of organic anions given in this section on Canada, therefore, have an apparent uncertainty of up to  $\pm 50\%$ . The acid functional group content of lakes from the Eastern Lake Survey was estimated to be  $12 \mu\text{eq} (\text{mg DOC})^{-1}$  (Driscoll et al. 1989). The median organic anion concentration is between 25 and  $60 \mu\text{eq L}^{-1}$  for each region in southeastern Canada (Fig. 10). Higher median values of organic anions occur in North Ontario, West Ontario, and the North Atlantic, and the lowest median value occurs in Sudbury/Noranda.

Lakes having ANC levels of  $\leq 50 \mu\text{eq L}^{-1}$  exhibit the same general geographic pattern in major ion chemistry (Fig. 11) as those with  $\text{ANC} \leq 400 \mu\text{eq L}^{-1}$ . From an acid-base chemistry perspective, the reason for the difference in median ANC for these two ANC strata is that, within any region, the lakes with  $\text{ANC} \leq 50 \mu\text{eq L}^{-1}$  have lower median  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  than lakes with  $\text{ANC} \leq 400 \mu\text{eq L}^{-1}$  (Figs. 10 and 11). Intra-regional differences in lake-water ANC for these two ANC strata are, therefore, primarily because of differences in base cation concentrations, which are the result of intraregional differences in bedrock and soil geochemistry.

The role of spatial variation in lake-water  $\text{SO}_4^{2-}$  in explaining the variation in ANC is secondary relative to that of base cation concentrations. Within any region, the 25th, 50th, and 75th percentiles of  $\text{SO}_4^{2-}$  concentrations of the lakes with  $\text{ANC} \leq 400 \mu\text{eq L}^{-1}$  and lakes with  $\text{ANC} \leq 50 \mu\text{eq L}^{-1}$  are similar. However, because the lakes having  $\text{ANC} \leq 50 \mu\text{eq L}^{-1}$  have base cation concentrations that are similar to  $\text{SO}_4^{2-}$  concentrations (Fig. 11),  $\text{SO}_4^{2-}$  has a greater influence on the acid-base chemistry. The observation that median and interquartile ranges of lake-water  $\text{SO}_4^{2-}$  are similar for the two ANC strata indicates that, within each region, most lakes with  $\text{ANC} \leq 400 \mu\text{eq L}^{-1}$  have a relatively uniform  $\text{SO}_4^{2-}$  concentration (Figs. 10 and 11). Furthermore, this similarity suggests that sulfate is not a major influence on within region ANC variability. The within-region variability in lake-water  $\text{SO}_4^{2-}$  (approximate interquartile range:  $30 \mu\text{eq L}^{-1}$ ) is likely because of variability in wet and dry atmospheric deposition of  $\text{SO}_4^{2-}$ .

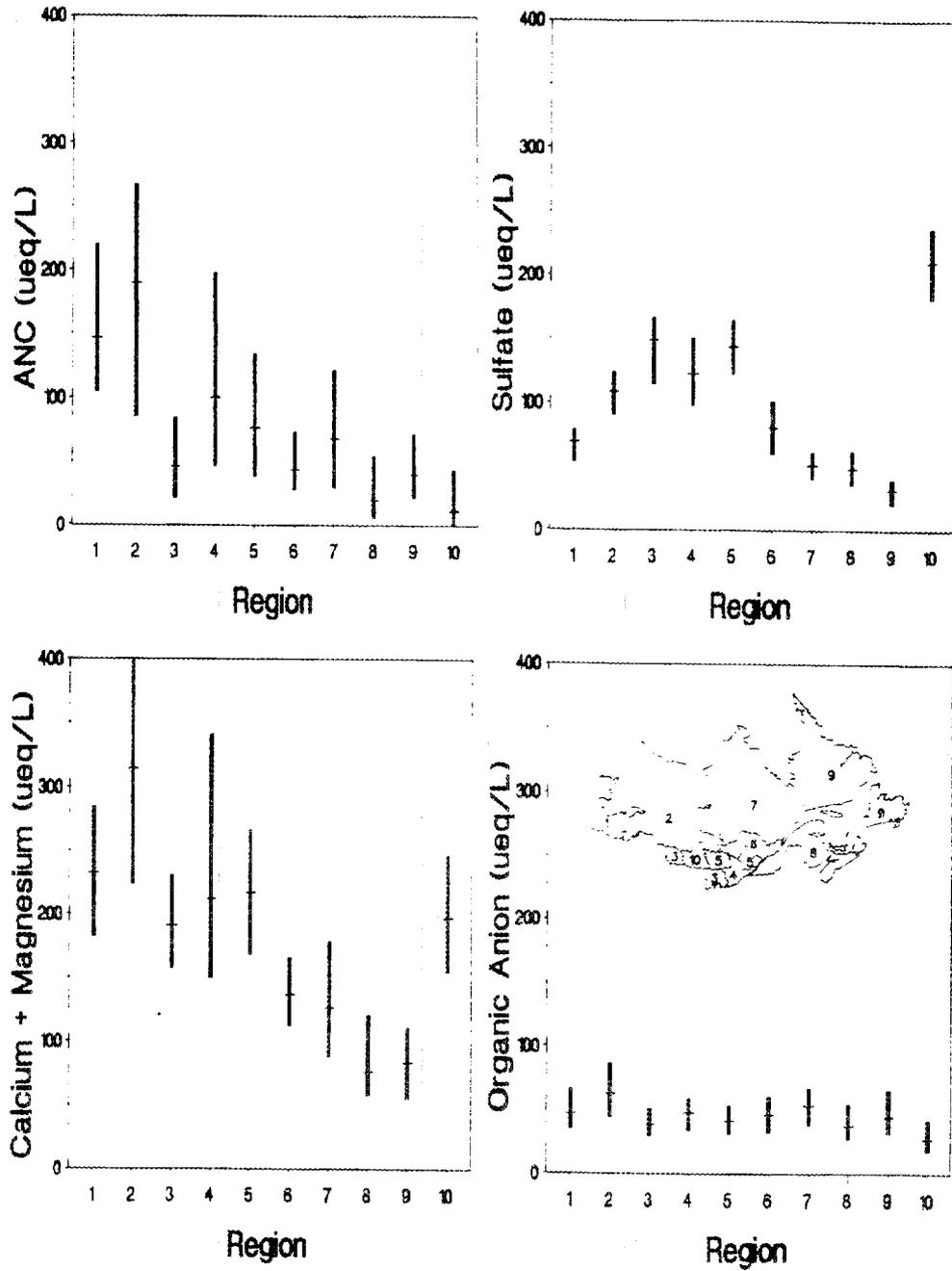


Fig. 10. Medians and quartiles of ANC, sulfate, calcium plus magnesium, and organic anion by region, based on Level 3 sampled lakes with  $\text{ANC} \leq 400 \mu\text{eq L}^{-1}$ . See Fig. 1 and Table 2 for the definition of regions.

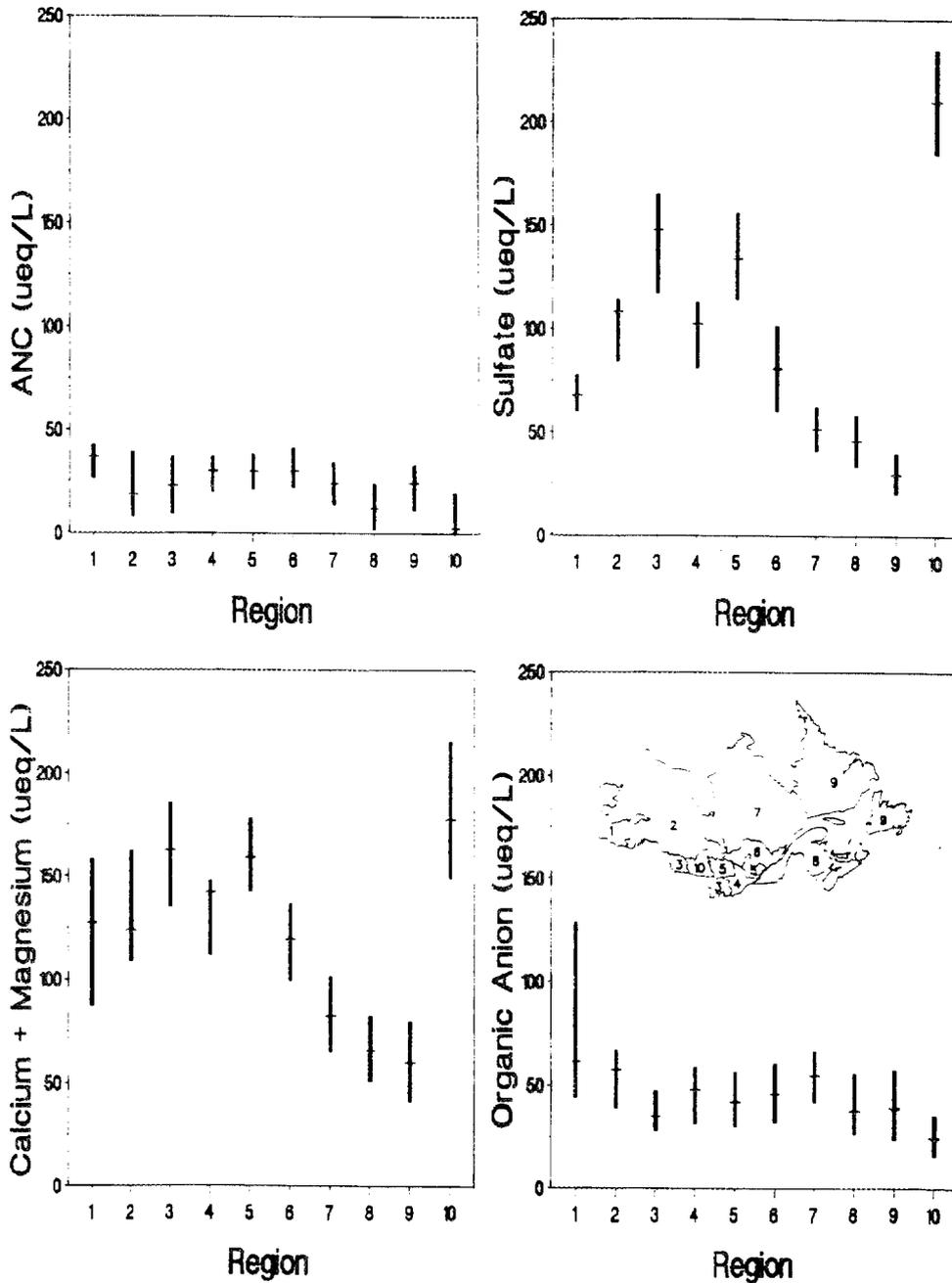


Fig. 11. Medians and quartiles of ANC, sulfate, calcium plus magnesium, and organic anion by region, based on Level 3 sampled lakes having  $\text{ANC} \leq 50 \mu\text{eq L}^{-1}$ . See Fig. 1 and Table 2 for the definition of regions.

and in the biogeochemistry of  $\text{SO}_4^{2-}$  in watersheds (Cook and Jager, in press).

### 3.3 Relationships Between Lake-Water $\text{SO}_4^{2-}$ and $\text{SO}_4^{2-}$ Deposition

Analysis of data in the United States and in Scandinavia has shown a strong relationship between wet  $\text{SO}_4^{2-}$  deposition and surface water  $\text{SO}_4^{2-}$  (Sullivan et al. 1988, Kaufmann et al. 1988, Wright and Henriksen 1978, Baker et al. 1990). For southeastern Canada, the lake-water  $\text{SO}_4^{2-}$  and wet  $\text{SO}_4^{2-}$  deposition (Fig. 12) also showed a significant relationship. The correlation coefficient for the regional median values shown in Fig. 12 was 0.79 ( $p < 0.01$ ; weighted by the reciprocal of the interquartile range). The implication from this plot is that for many lakes in southeastern Canada, atmospheric  $\text{SO}_4^{2-}$  deposition is the major control on lake-water  $\text{SO}_4^{2-}$  concentrations. Evidence from input-output mass balance studies of individual watersheds on the Precambrian Shield in southeastern Canada shows that atmospheric deposition is the primary source of  $\text{SO}_4^{2-}$  (Schindler et al. 1976, Jeffries et al. 1988, Nicholson 1988, LaZerte and Dillon 1984).

The variability in the data in Fig. 12 is not surprising because local variation in deposition was not incorporated, estimates of dry deposition were not included, and we did not control for other sources of variability (regional geology and hydrology, morphology of the drainage basins, and lake bathymetry). For example, differences in hydrology among lakes receiving the same deposition will result in differences in lake-water  $\text{SO}_4^{2-}$  (Kelly et al. 1987, Baker and Brezonik 1988). Without further information, the relative importance of wet deposition, dry deposition, regional geology and hydrology, basin morphology, and lake bathymetry in accounting for the variability in Fig. 12 cannot be determined.

### 3.4 Relationships Between Lake-Water Chemistry Variables and Lake-Water $\text{SO}_4^{2-}$

#### 3.4.1 Plot Analysis

If  $\text{SO}_4^{2-}$  deposition has affected the acid-base chemistry of lakes, the observed spatial variability in current acid-base chemistry may possibly be explained by current  $\text{SO}_4^{2-}$  deposition levels, with higher concentrations of lake-water  $\text{SO}_4^{2-}$  associated with lower values of ANC (W. Warnick, Department of Energy, personal communication, September 1988). The degree of such associations with lake-water chemistry were investigated by analyzing plots of ANC or  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  against lake-water  $\text{SO}_4^{2-}$  for Level 3 lakes for the nine regions (Figs. 13, 14, 15, and 16) and for Sudbury/Noranda (Fig. 17). In Figs. 14 and 15, the nine plots are generally arranged such that the northern regions are at the top of each set, western regions on the left, and eastern regions on the right. Lake-water  $\text{SO}_4^{2-}$  is used to represent  $\text{SO}_4^{2-}$  deposition because of the difficulties in estimating total  $\text{SO}_4^{2-}$  deposited for individual lakes (see Sect. 1.2.3).

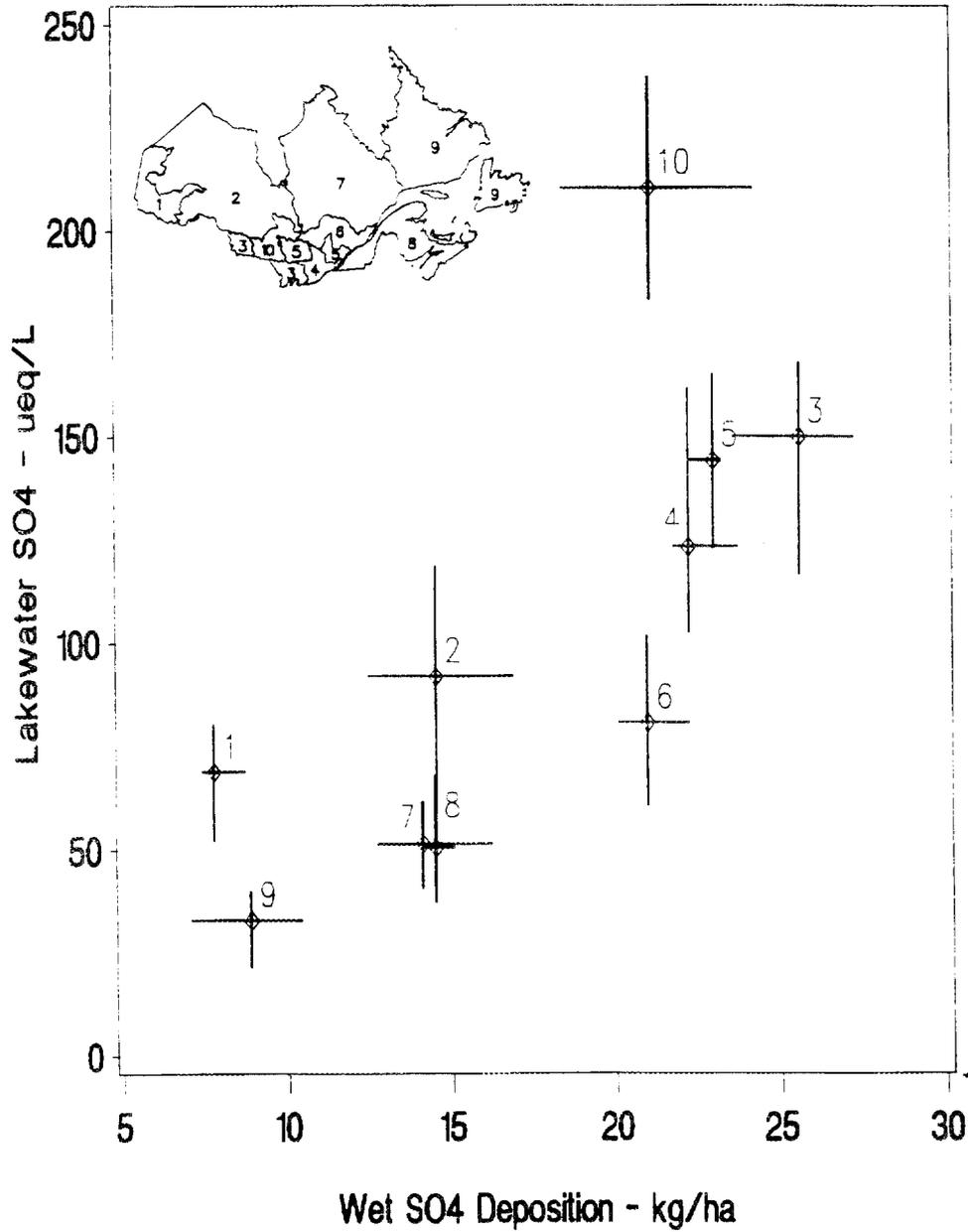


Fig. 12. Medians and quartiles of lake-water  $\text{SO}_4^{2-}$  ( $\mu\text{eq L}^{-1}$ ) vs wet  $\text{SO}_4^{2-}$  deposition (1982-1986) annual average in  $\text{g/m}^2$ ) by region for Level 1 sampled lakes in southeastern Canada. See Fig. 1 and Table 2 for the definition of regions.

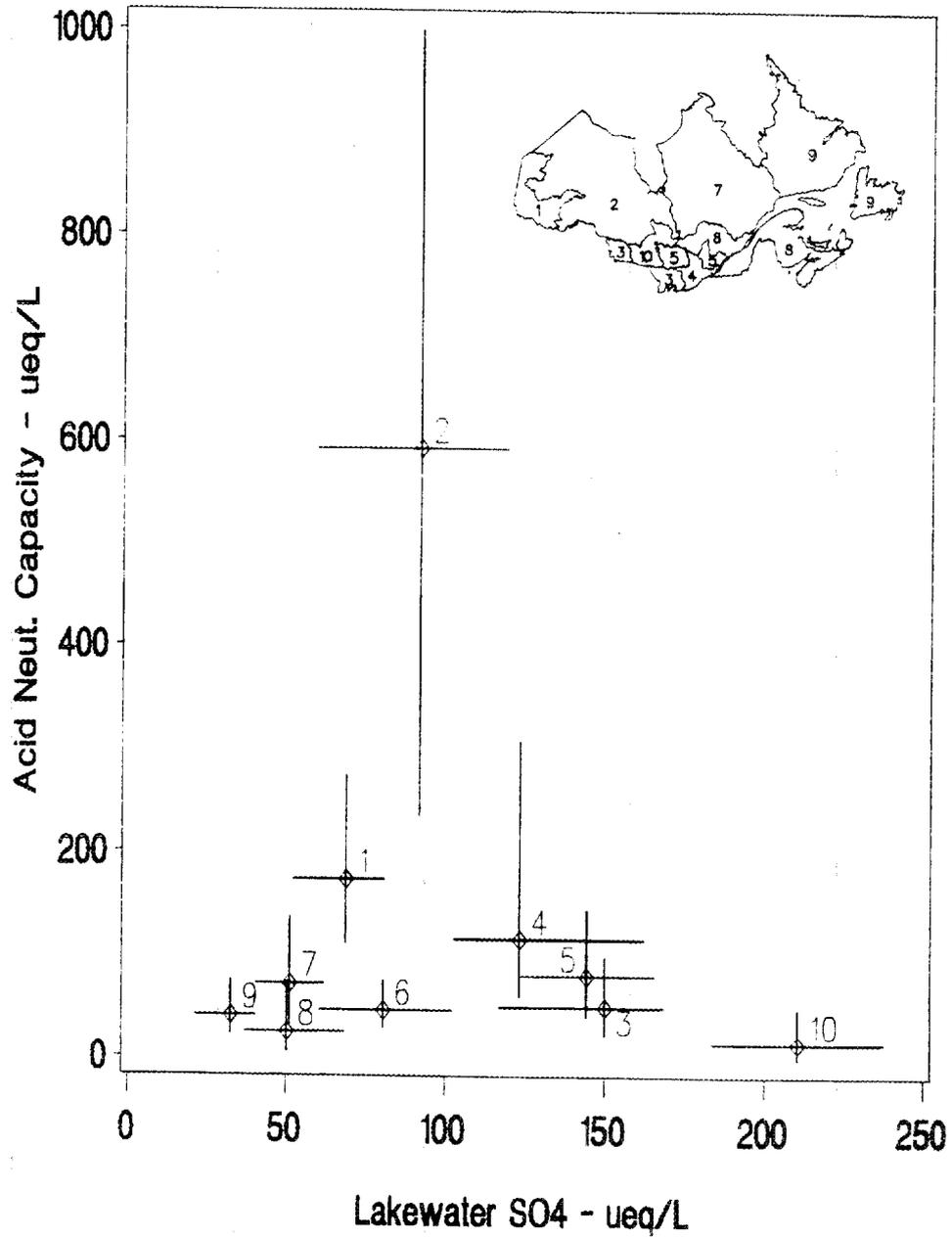


Fig. 13. Medians and quartiles of ANC vs  $\text{SO}_4^{2-}$  by region for Level 3 sampled lakes in southeastern Canada. See Fig. 1 and Table 2 for the definition of regions.

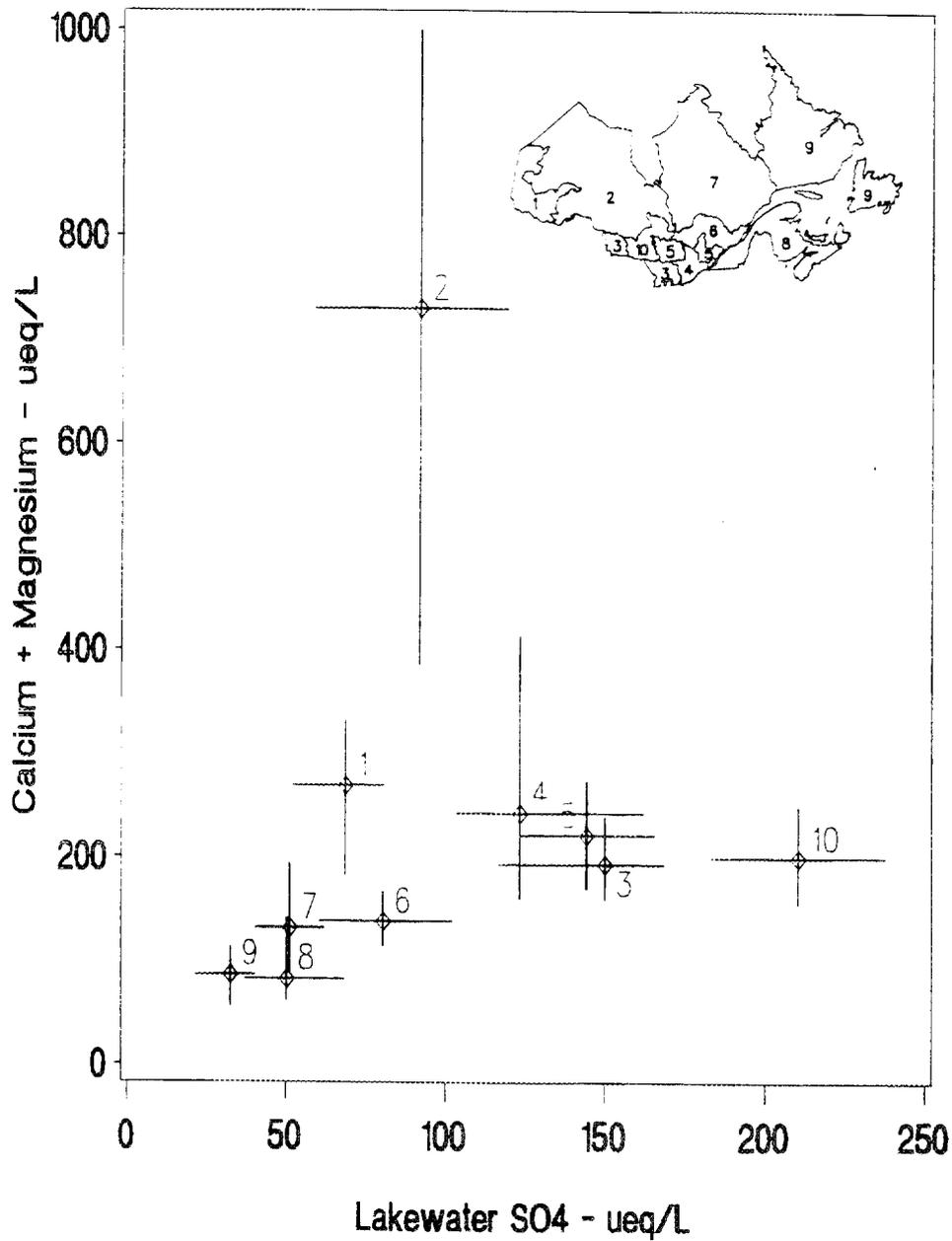


Fig. 14. Medians and quartiles of calcium plus magnesium [ $\text{Ca}^{2+} + \text{Mg}^{2+}$ ] vs  $\text{SO}_4^{2-}$  by region for Level 3 sampled lakes in southeastern Canada. See Fig. 1 and Table 2 for the definition of regions.

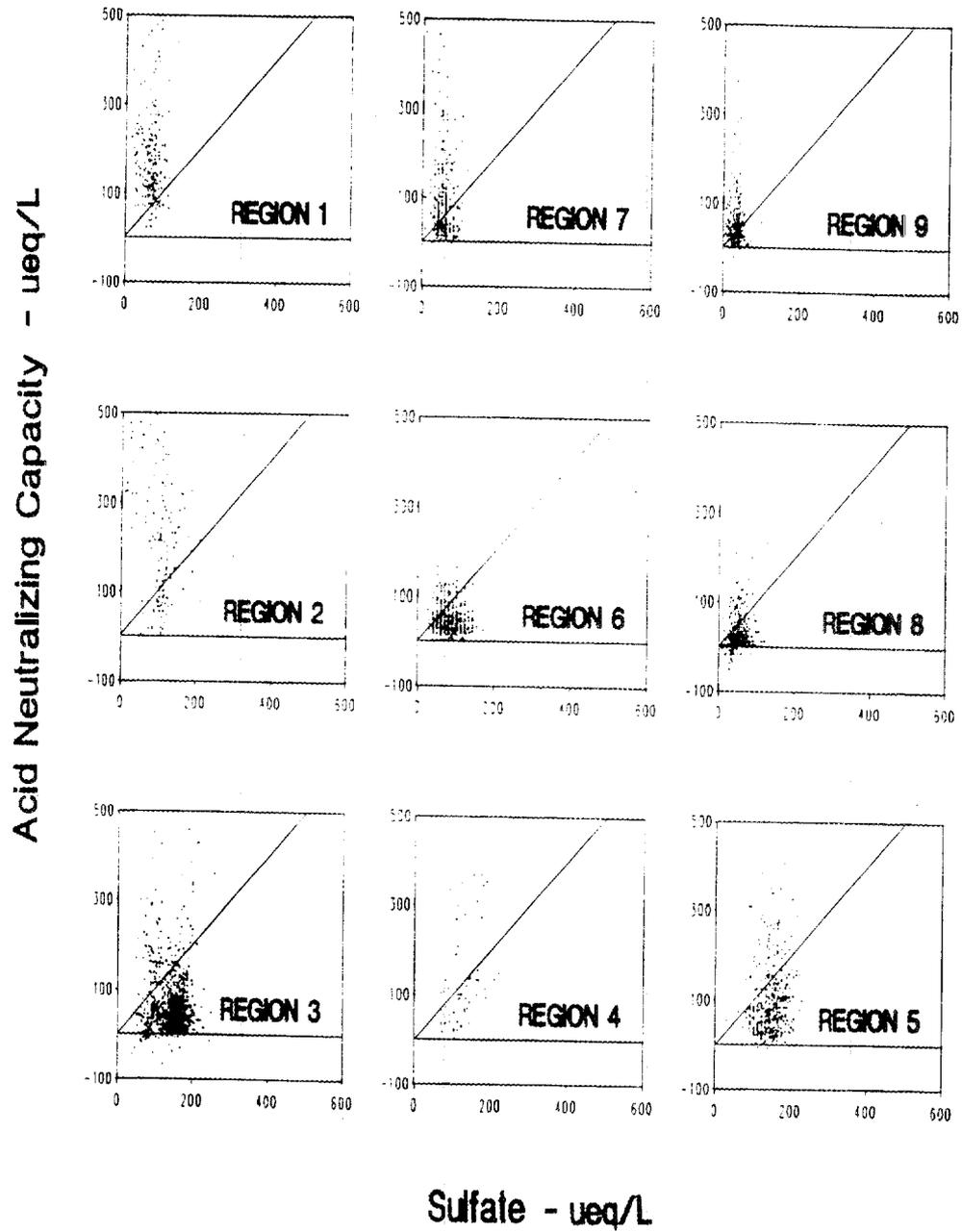


Fig. 15. ANC vs  $\text{SO}_4^{2-}$  for Level 3 sampled lakes in southeastern Canada by regions. Reference line represents a 1:1 relationship.

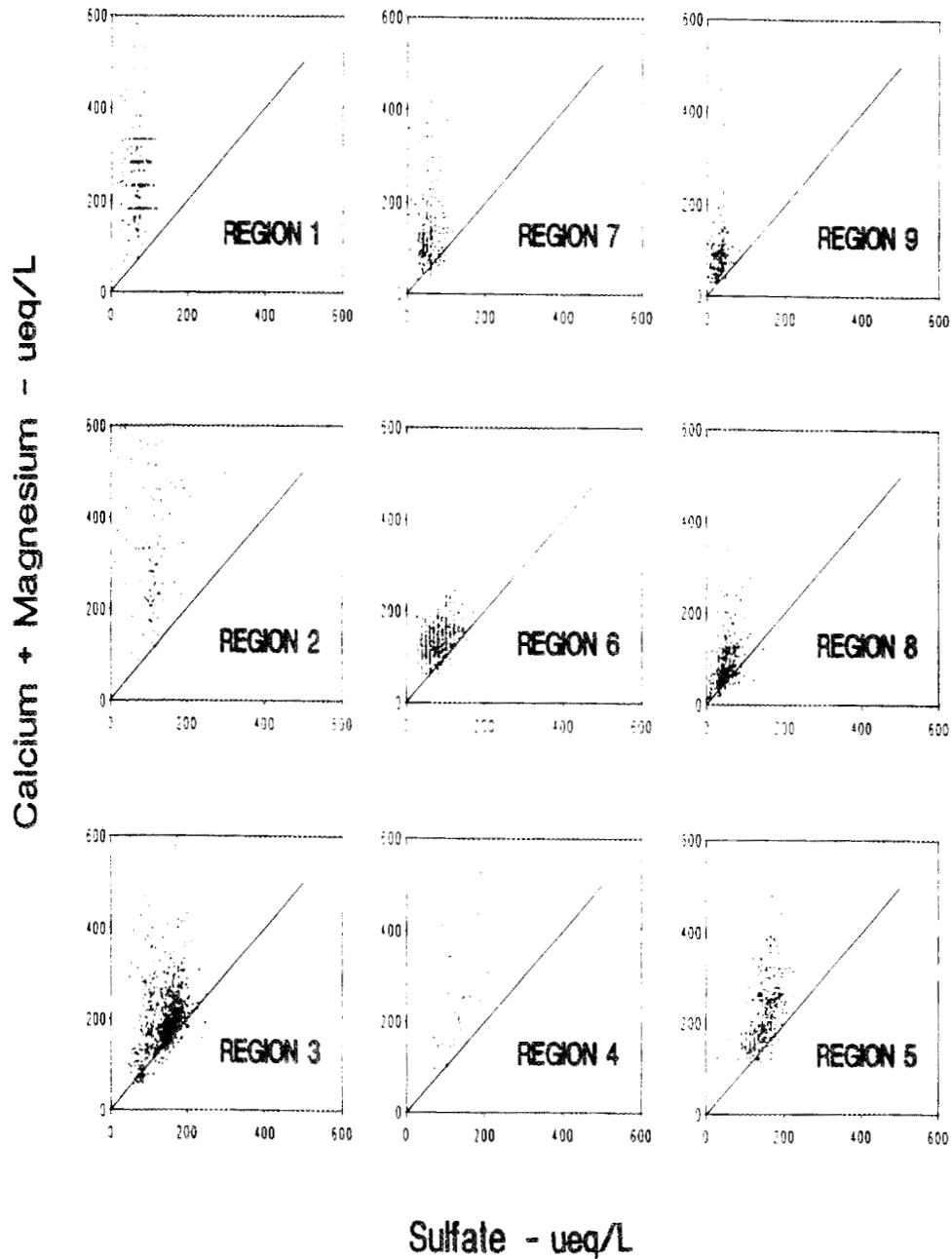


Fig. 16.  $[Ca^{2+} + Mg^{2+}]$  vs  $SO_4^{2-}$  for Level 3 sampled lakes in southeastern Canada by regions. Reference line represents a 1:1 relationship.

## Region 10 - Sudbury

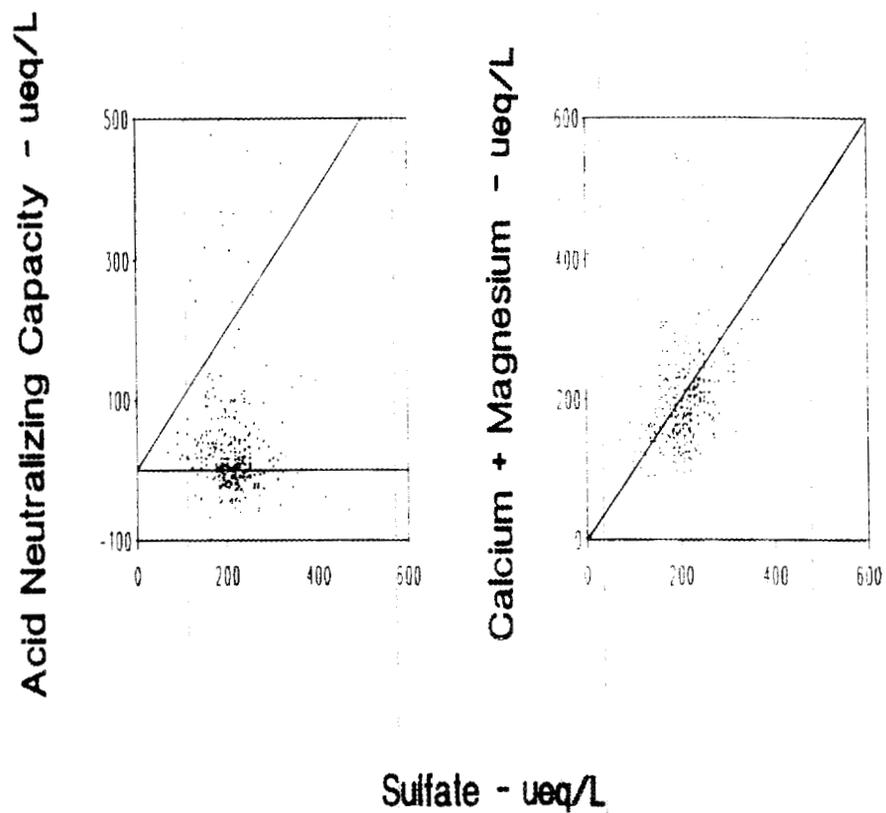


Fig. 17. ANC vs  $\text{SO}_4^{2-}$  and  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  vs  $\text{SO}_4^{2-}$  for Level 3 sampled lakes in the Sudbury/Noranda region. Reference line represents a 1:1 relationship.

These plots show a great deal of variability. For a relatively narrow range of  $\text{SO}_4^{2-}$  (25 to 250  $\mu\text{eq L}^{-1}$ ), ANC and  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  exhibit a large range (0 to 1000  $\mu\text{eq L}^{-1}$ ). If  $\text{SO}_4^{2-}$  were causing a loss of ANC in which the  $\text{SO}_4^{2-}$  replaced ANC and if all other factors were constant, there should be a negative correlation between the two variables (Figs. 13, 15, and 17a). The absence of a strong relationship between  $\text{SO}_4^{2-}$  and ANC and  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  indicates that other factors are important in controlling the lake-water concentrations of ANC and  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  (e.g., other factors were not constant). These other factors include bedrock and soil chemistry, soil thickness, and hydrologic flowpath and are discussed in detail in Turner et al. (1990). Sect. 1.2.5 examines the underlying relationships that may be contributing to these patterns by stratifying the population of sampled lakes and considering the acid-base chemistry. A better approach for examination of the relationship between the acid-base chemistry and atmospheric deposition would be to plot directly the change in ANC over the past 100 years as a function of the change in  $\text{SO}_4^{2-}$  deposition over that time period (Sullivan et al. 1988); data from Canadian lakes for this type of plot do not exist.

### 3.4.2 Interaction of Atmospheric Deposition and Organic Acids

Atmospheric deposition may also cause a reduction in the concentration of DOC and organic acids through a change in biogeochemical cycling of organic compounds or through direct chemical reaction of  $\text{H}^+$  with organic anions or dissolved organic carbon, for example, aggregation and flocculation (Turner et al. 1990, Tipping and Ohnstad 1984). At Plastic Lake during a 6-year period (1980–1985), Dillon et al. (1987) observed a decrease in DOC and organic anion concentrations with decreasing ANC. The cause of the acidification was attributed to low base cation leaching not  $\text{SO}_4^{2-}$  deposition, which actually decreased during this period.

To investigate the interaction between atmospheric deposition and organic acids, we plotted lake-water  $\text{SO}_4^{2-}$  and organic anion for all of the regions in southeastern Canada (Fig. 18). Lake-water  $\text{SO}_4^{2-}$  was chosen to represent atmospheric deposition because of the difficulties in estimating total deposition, as described in the preceding paragraphs.

The  $\text{SO}_4^{2-}$  and organic anion data follow a weak inverse relationship, with high organic anion associated with low  $\text{SO}_4^{2-}$  and high  $\text{SO}_4^{2-}$  associated with low organic anion (Fig. 18). The data are consistent with the hypothesis that increases in atmospheric deposition caused organic anion concentrations to decline. However, a number of alternative hypotheses may also explain this inverse relationship between organic anion and  $\text{SO}_4^{2-}$ . For example, the location of the lakes in southeastern Canada may contribute to the existence of an inverse relationship between atmospheric deposition (Fig. 2) and percentage wetlands (Fig. 4). Wetlands are more abundant in West and North Ontario, areas that receive relatively low deposition, and wetlands are less abundant in southeastern Ontario, an area that receives relatively high deposition. Further analysis of the data in Fig. 18 reveals that the majority of high organic anion and low  $\text{SO}_4^{2-}$

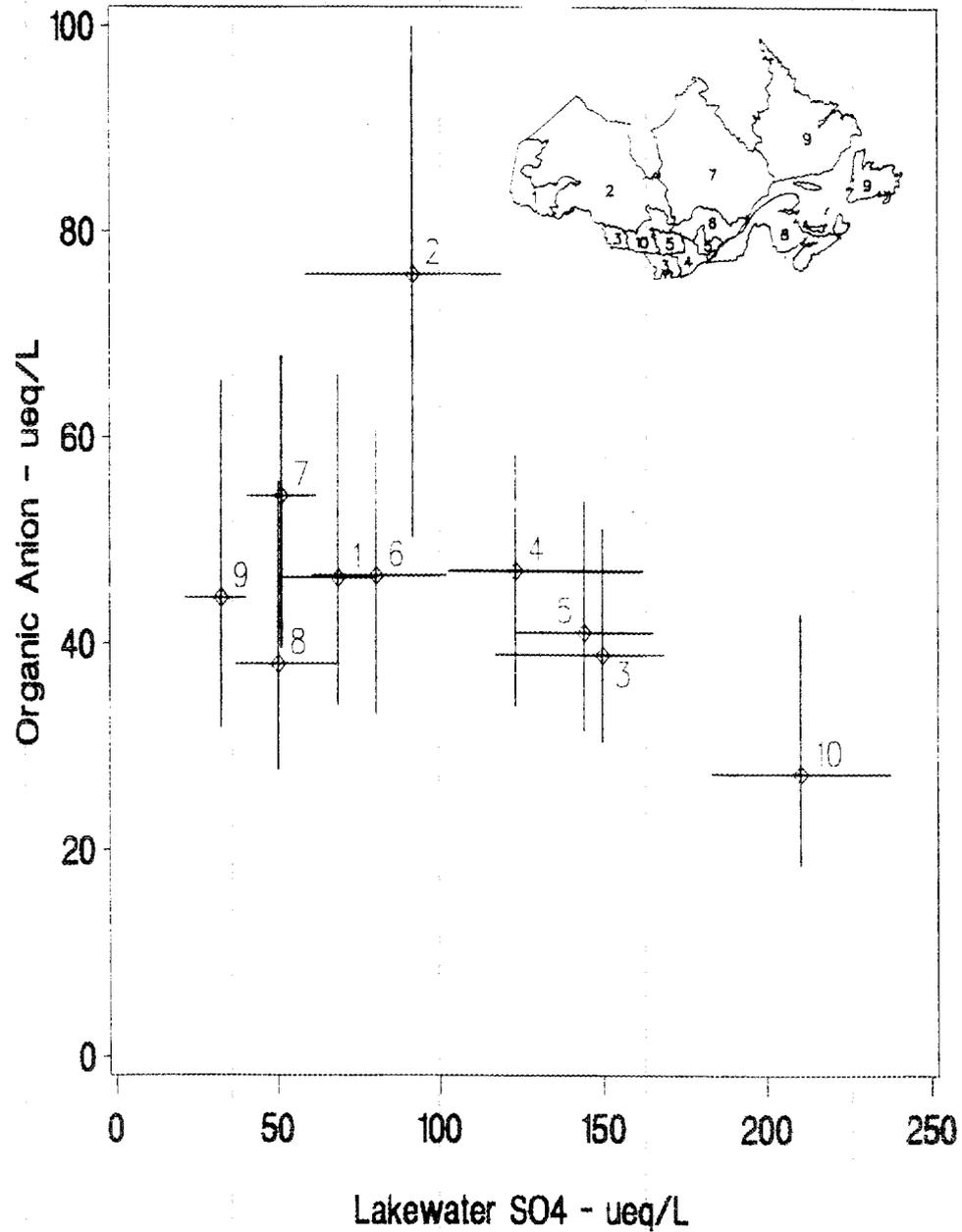


Fig. 18. Medians and quartiles of organic anion vs  $\text{SO}_4^{2-}$  by region for Level 3 sampled lakes in southeastern Canada. Organic anion was estimated using Oliver et al. (1983). See Fig. 1 and Table 2 for the definition of regions.

lakes were either in West Ontario or North Ontario; these organic-dominated lakes in North Ontario and West Ontario generally have ANC  $>0 \mu\text{eq L}^{-1}$ .

Another hypothesis that may explain the weak inverse relationship in Fig. 18 deals with the biogeochemical cycling of organic anion and  $\text{SO}_4^{2-}$ . Wetlands and riparian zones may retain  $\text{SO}_4^{2-}$  through bacterial sulfate reduction and also may produce organic anions through the partial decomposition of organic matter (Turner et al. 1990, Hemond in press). This hypothesis may explain the variation in organic anions and  $\text{SO}_4^{2-}$  within regions that receive relatively constant  $\text{SO}_4^{2-}$  from deposition. Without additional data, the relative importance of each of these three possible mechanisms in explaining the relationships in Fig. 18 cannot be determined.

### 3.5 Bivariate Relationships Among Lake-water Chemistry Variables

The major ion chemistry of surface waters represents the integration of atmospheric deposition and a number of biogeochemical processes occurring in watersheds and lakes (Turner et al. 1990). Associations between variables derived from synoptic surveys of lake-water chemistry provide a means of qualitatively estimating the processes responsible for lake-water chemistry. The following plots were made for Level 3 lakes having ANC  $\leq 400 \mu\text{eq L}^{-1}$ . Level 3 data were used for these analyses to ensure that observations had the requisite solute data (ANC, pH,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ , and  $\text{K}^+$ ).

#### 3.5.1 ANC vs pH

Plots of pH vs ANC for regional lake-water chemistry data may be used to examine the influence of  $\text{CO}_2$ , weak organic acids, and aluminum on pH measurements. If carbonate species ( $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$ ) and  $\text{H}^+$  comprise the lake-water ANC (e.g.,  $\text{ANC} = \text{HCO}_3^- + \text{CO}_3^{2-} - \text{H}^+$ ) and if the lake water is in equilibrium with atmospheric  $\text{CO}_2$ , the pH and ANC data should fall on the theoretical line (Stumm and Morgan 1980) in Fig. 19. For all the regions in southeastern Canada, the data generally fall below the theoretical curve for ANC concentrations  $>50 \mu\text{eq L}^{-1}$ . This deviation of data from the theoretical curve is caused by weak acids (e.g., elevated partial pressures of  $\text{CO}_2$ , organic acids, and aqueous aluminum) that promote lower pH values than that expected from a system in equilibrium with atmospheric  $\text{CO}_2$  and only containing carbonate species.

Elevated partial pressures of  $\text{CO}_2$ , produced in the lake by respiration or stream or groundwater input, have been observed in other regional surveys (Cook and Jager in press, Elwood et al. in press). The influence of elevated partial pressures of  $\text{CO}_2$  may be determined by measuring air-equilibrated pH, which is not available for the Canadian data base. For the surveys in southeastern Canada, precautions were not specifically taken against change in  $\text{CO}_2$  content of the lake-water samples. Therefore, the partial pressure of  $\text{CO}_2$  may range from in situ values to equilibrium with atmospheric partial pressures. For a given ANC, elevated levels of  $\text{CO}_2$  cause a lower pH

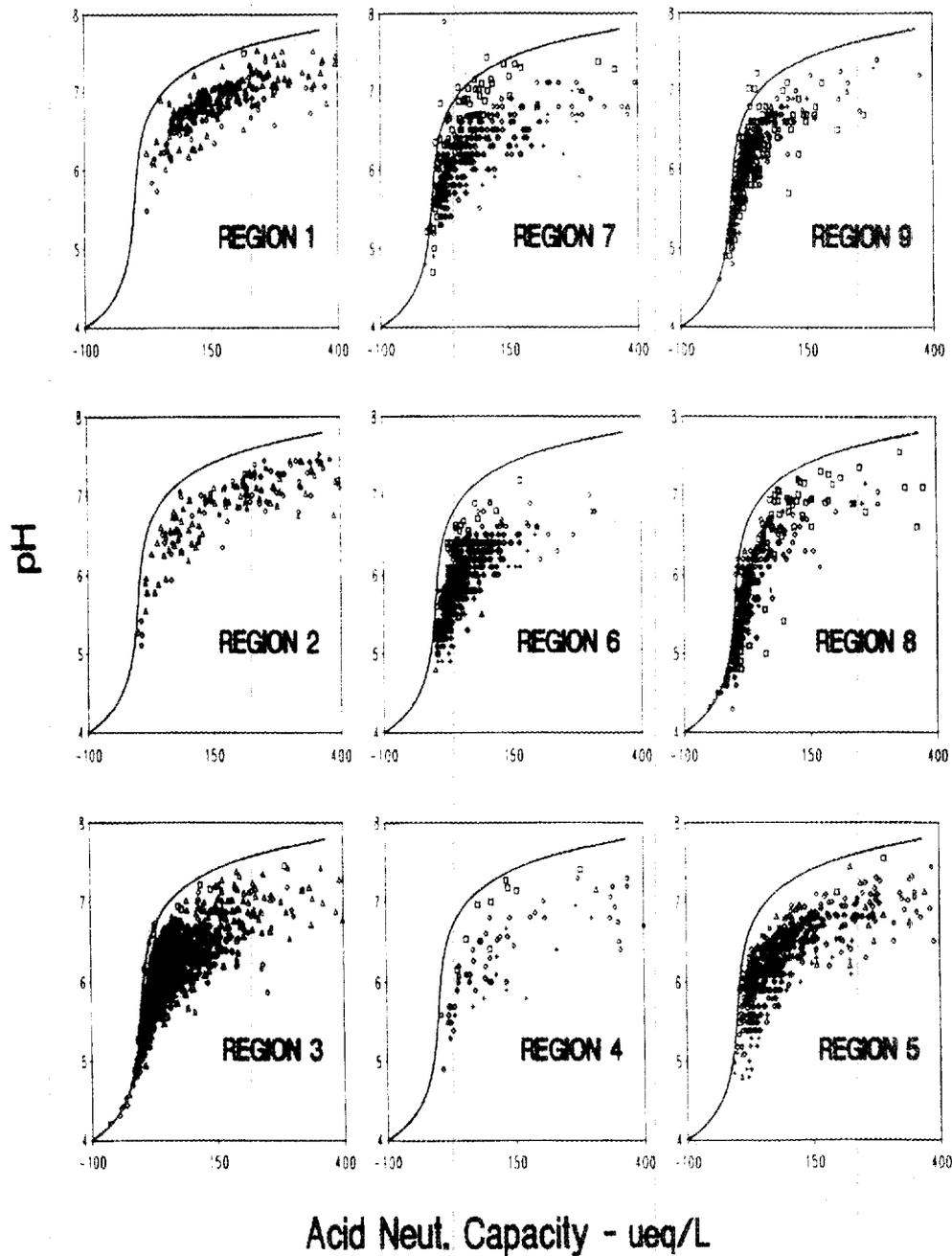


Fig. 19. pH vs ANC for Level 3 sampled lakes in southeastern Canada by regions. Reference line represents theoretical pH-ANC relationship for lake water in equilibrium with atmospheric CO<sub>2</sub> (Stumm and Morgan 1980). Symbols indicate the DOC ( $\mu\text{mol L}^{-1}$ ) level as follows: square =  $\leq 200$ ; triangle = 200 to  $\leq 600$ ; diamond =  $> 600$ ; and plus = missing data.

relative to that of a sample at equilibrium with atmospheric  $\text{CO}_2$  (Stumm and Morgan 1980).

Organic acids may also cause a lower pH for any given value of ANC because organic acids [average dissociation constants ( $\text{pK} = 3.5$ )] are somewhat stronger acids than carbonic acid ( $\text{pK} = 6.3$ ) (Cook and Jager in press, Turner et al. 1990). The influence of aluminum on the pH-ANC relationship should be seen at low pH ( $\text{pH} < 5.0$ ), where aluminum may be present in greater concentrations (Munson and Gherini, in press). Good agreement between the data and the theoretical curve for low pH values suggests that the acid-base chemistry of aluminum is not affecting the pH-ANC relationship greatly.

Without a method for separating the effect of  $\text{CO}_2$  and organic acids on the pH and ANC relationship, the exact cause of the deviation in Fig. 19 cannot be determined. The data indicate that pH is lower for the lake-water samples than that expected for a  $\text{HCO}_3^-$  solution at the same ANC concentration and in equilibrium with atmospheric  $\text{CO}_2$ .

### 3.5.2 Major Ion Chemistry

In areas such as the Western United States where ANC is the primary anion in surface waters and the concentrations of other ions ( $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{Al}^{3+}$ ,  $\text{K}^+$ , and  $\text{Na}^+$ ) are low, a plot of ANC vs  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  follows the  $\text{ANC} = [\text{Ca}^{2+} + \text{Mg}^{2+}]$  line (Melack and Stoddard in press). In regions where the data fall on this line, base cation production via weathering and cation exchange is driven by carbonic acid, with ANC ( $\text{HCO}_3^-$  and  $\text{A}^-$ ),  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  as the major products (Stumm and Morgan 1980). In southeastern Canada, the data for Regions 1, 7, and 9 fall near the line, indicating that the lake water is predominantly a solution of base cations and ANC for  $\text{ANC} > 50 \mu\text{eq L}^{-1}$  (Fig. 20). These regions receive low levels of atmospheric deposition (Table 2). Data for the other regions in southeastern Canada generally fall below the  $\text{ANC} = [\text{Ca}^{2+} + \text{Mg}^{2+}]$  line (Figs. 20 and 21), indicating that other anions contribute to the charge balance.

The additional contribution of  $\text{SO}_4^{2-}$  to the anionic charge may be shown with a plot of  $[\text{ANC} + \text{SO}_4^{2-}]$  against  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  (Figs. 21 and 22), which yields a better fit to the 1:1 line than the data in Fig. 20. The implication from this improved fit is that the other ions ( $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Cl}^-$ ) are of relatively minor importance or that the charge contribution of cations not in the figure is balanced by that of anions not in the figure.

The observation that  $\text{SO}_4^{2-}$  contributes significantly to the anionic charge suggests that either (1) sulfuric acid weathering reactions contribute to base cation production, with the source of sulfuric acid being either atmospheric deposition or reduced sulfur in bedrock; (2) there are sources of neutral salts of  $\text{SO}_4^{2-}$  (e.g.,  $\text{CaSO}_4$ ) in the regions; (3) acidification has occurred, with ANC lost and  $\text{SO}_4^{2-}$  replacing the lost ANC; or (4) some combination of these three. Sources of sulfur, either neutral salts or reduced sulfur, may occur in the Precambrian bedrock and Pleistocene till in southeastern Canada (Regions 3, 4, 5, and 10); these sources may contribute to the relatively high lake-water  $\text{SO}_4^{2-}$  concentrations ( $\text{SO}_4^{2-} > 200 \mu\text{eq L}^{-1}$ ) in Fig. 12. In addition, elevated local atmospheric deposition near

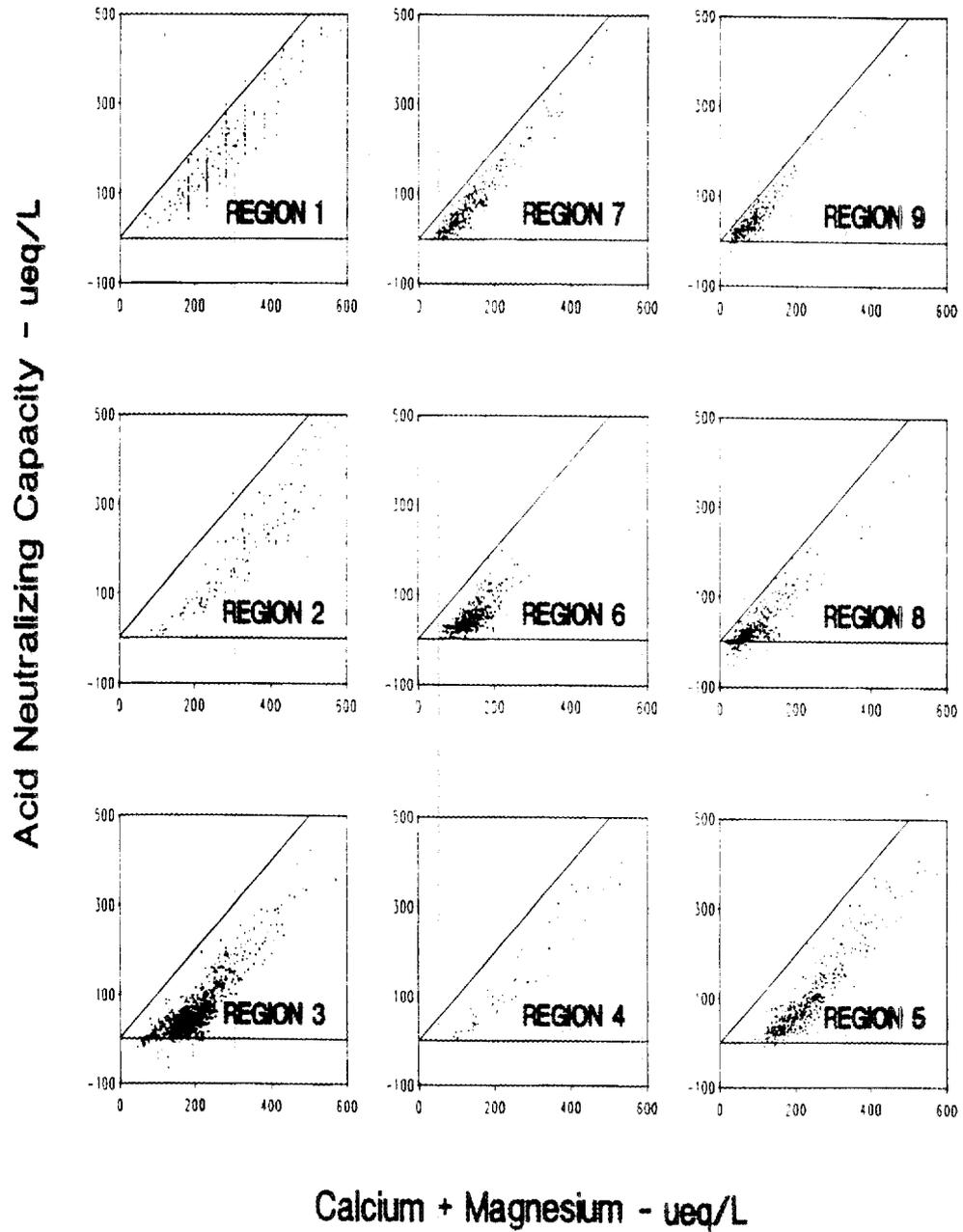


Fig. 20. ANC vs  $[Ca^{2+} + Mg^{2+}]$  for Level 3 sampled lakes in southeastern Canada by regions. Reference line represents a 1:1 relationship.

## Region 10 - Sudbury

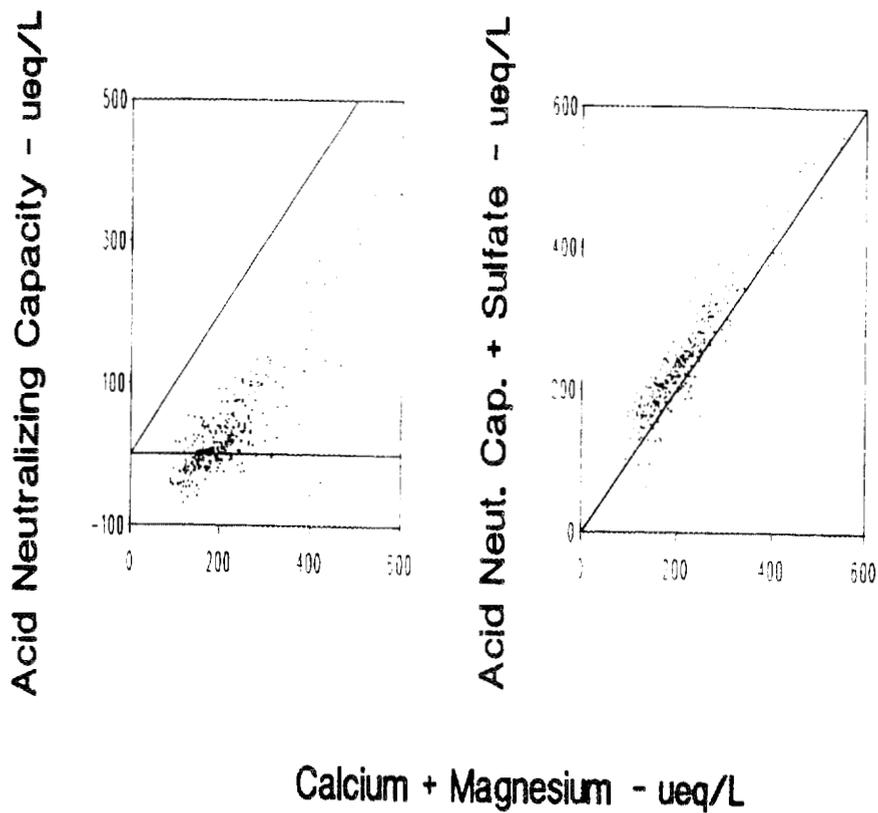


Fig. 21. ANC vs  $[Ca^{2+} + Mg^{2+}]$  and ANC +  $SO_4^{2-}$  vs  $[Ca^{2+} + Mg^{2+}]$  for Level 3 sampled lakes in the Sudbury/Noranda region. Reference line represents a 1:1 relationship.

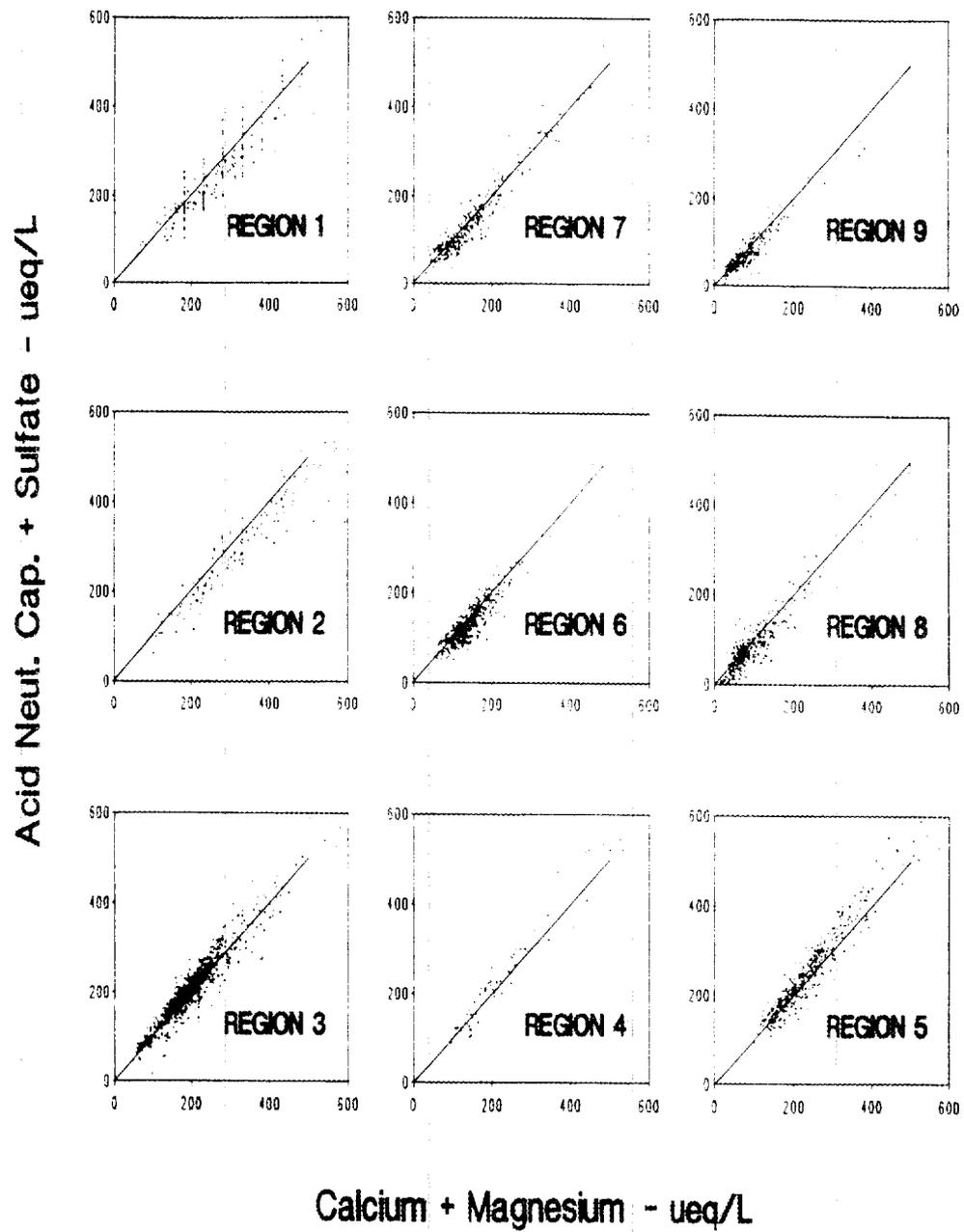


Fig. 22. ANC +  $\text{SO}_4^{2-}$  vs  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  for Level 3 sampled lakes in southeastern Canada by regions. Reference line represents a 1:1 relationship.

smelting and sintering activity may result in high concentrations of lake-water  $\text{SO}_4^{2-}$ . For Level 3 lakes with  $\text{SO}_4^{2-} \leq 200 \mu\text{eq L}^{-1}$  and for the data in the lake chemistry data base, we cannot distinguish among the first three causes. However, because the lake-water  $\text{SO}_4^{2-}$  concentrations for these lakes are all  $\leq 200 \mu\text{eq L}^{-1}$ , the majority of  $\text{SO}_4^{2-}$  is likely to be derived from the atmosphere, ruling out major in-watershed sources of sulfur. Intensive studies of watersheds on the Precambrian Shield in northwestern and southwestern Ontario reveal that lake-water  $\text{SO}_4^{2-}$  is derived primarily from atmospheric deposition (Schindler et al. 1976, Jeffries et al. 1988, Nicholson 1988, LaZerte and Dillon 1984). In addition, for lakes with  $\text{SO}_4^{2-} > [\text{Ca}^{2+} + \text{Mg}^{2+}]$  and with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$ ,  $\text{SO}_4^{2-}$  is not likely to be derived from neutral salts.

### 3.5.3 Relative Importance of $\text{SO}_4^{2-}$ and Organic Anion

The ratio  $\text{SO}_4^{2-} : [\text{Ca}^{2+} + \text{Mg}^{2+}]$  provides an estimate of the relative contribution of base cations and  $\text{SO}_4^{2-}$  in surface waters. This ratio increases in each of the regions of southeastern Canada as ANC decreases (Figs. 23 and 24). For lakes with high ANC, ANC (i.e., predominately  $\text{HCO}_3^-$ ) is the primary anion and  $\text{SO}_4^{2-}$  is a minor anion; values of  $\text{SO}_4^{2-} : [\text{Ca}^{2+} + \text{Mg}^{2+}]$  for high ANC systems should be low. For low-ANC lakes,  $\text{HCO}_3^-$  is a relatively minor anion and the importance of  $\text{SO}_4^{2-}$  in the charge balance is greater, as indicated by higher values of  $\text{SO}_4^{2-} : [\text{Ca}^{2+} + \text{Mg}^{2+}]$ . The observed increase in this ratio at low ANC is also the result of lower concentrations of  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  for relatively constant concentrations of  $\text{SO}_4^{2-}$  in low-ANC waters. Figs. 20 and 21 illustrate that lakes with low ANC typically have lower  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  than lakes with higher ANC.

Acidic lakes ( $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$ ) with  $\text{SO}_4^{2-}$  as the dominant ion at concentrations  $< 200 \mu\text{eq L}^{-1}$  have probably experienced acidification as a result of atmospheric deposition; these lakes are located in the upper left part of Figs. 23 and 24. Note that lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  in the Sudbury/Noranda region exhibit high values of  $\text{SO}_4^{2-} : [\text{Ca}^{2+} + \text{Mg}^{2+}]$ ; these lakes are acidic as the result of locally high atmospheric deposition originating from smelting activities. For acidic lakes having  $\text{SO}_4^{2-} : [\text{Ca}^{2+} + \text{Mg}^{2+}] \geq 1$ , rates of base cation production in the watershed-lake system are insufficient to neutralize inputs of acidity from atmospheric deposition or other sources. For lakes with ANC between 0 and  $100 \mu\text{eq L}^{-1}$ , this plot does not indicate whether the loss of ANC has or has not occurred; these lake-watershed systems may also have a low capacity to produce ANC.

The role of organic anions in the acid-base chemistry of surface waters can be evaluated from the concentration of organic anion and the ratio of organic anion to  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$ . A ratio greater than one indicates that the acidity of those lakes are substantially influenced by organic acids.

The organic anion:  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  ratio is generally low ( $< 0.5$ ) for most lakes having  $\text{ANC} > 100 \mu\text{eq L}^{-1}$  and increases to values greater than one for some systems in which  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  in the South Atlantic, North Atlantic, North Quebec, and southeast Ontario regions (Fig. 25). The South Atlantic and North Atlantic regions have the largest number

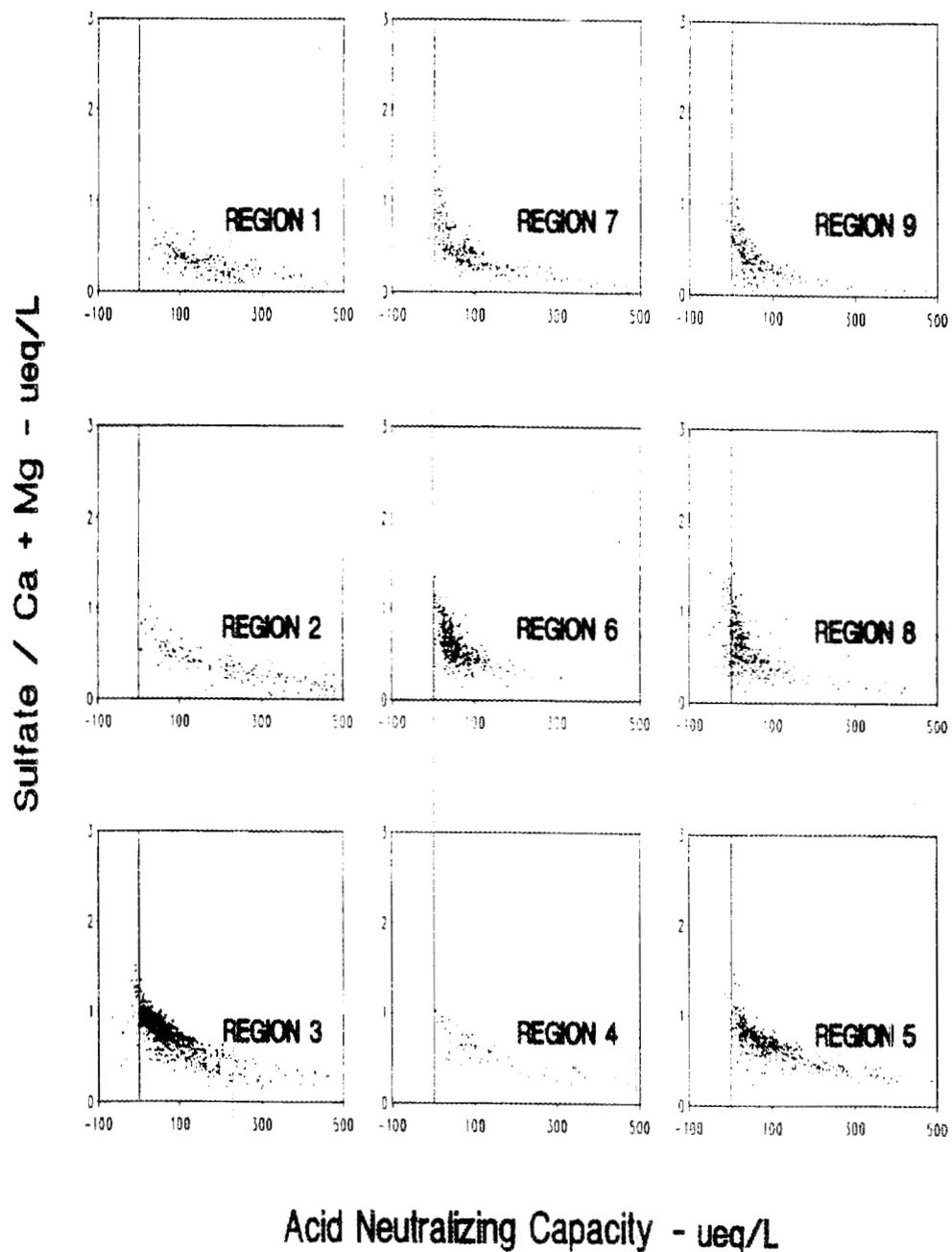


Fig. 23.  $\text{SO}_4^{2-}/[\text{Ca}^{2+} + \text{Mg}^{2+}]$  vs ANC for Level 3 sampled lakes in southeastern Canada by regions.

## Region 10 - Sudbury

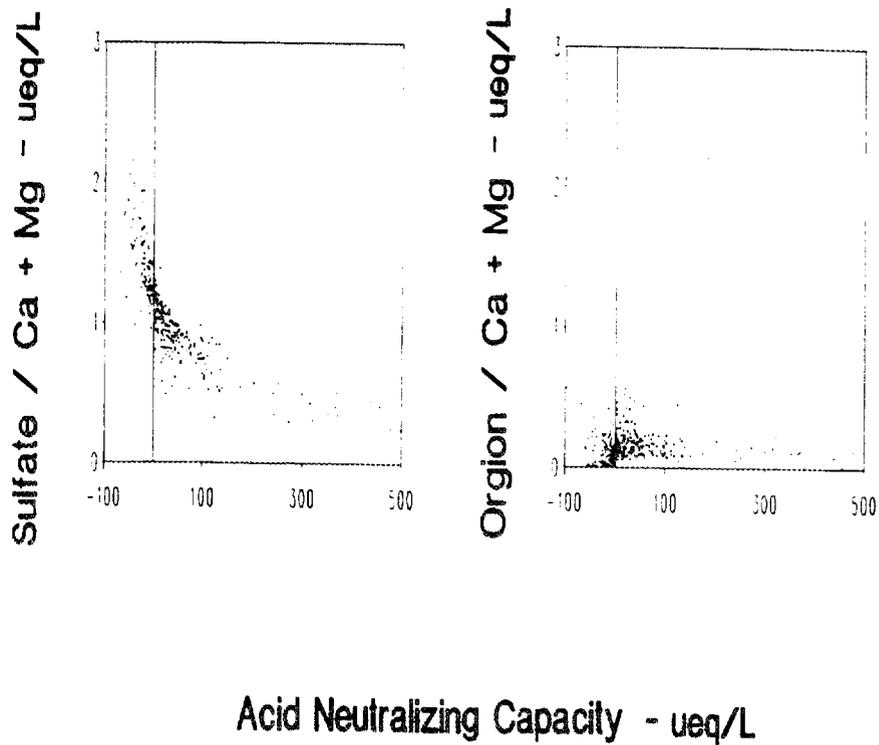


Fig. 24.  $\text{SO}_4^{2-}/[\text{Ca}^{2+} + \text{Mg}^{2+}]$  vs ANC and organic anion/ $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  vs ANC for Level 3 sampled lakes in the Sudbury/Noranda region.

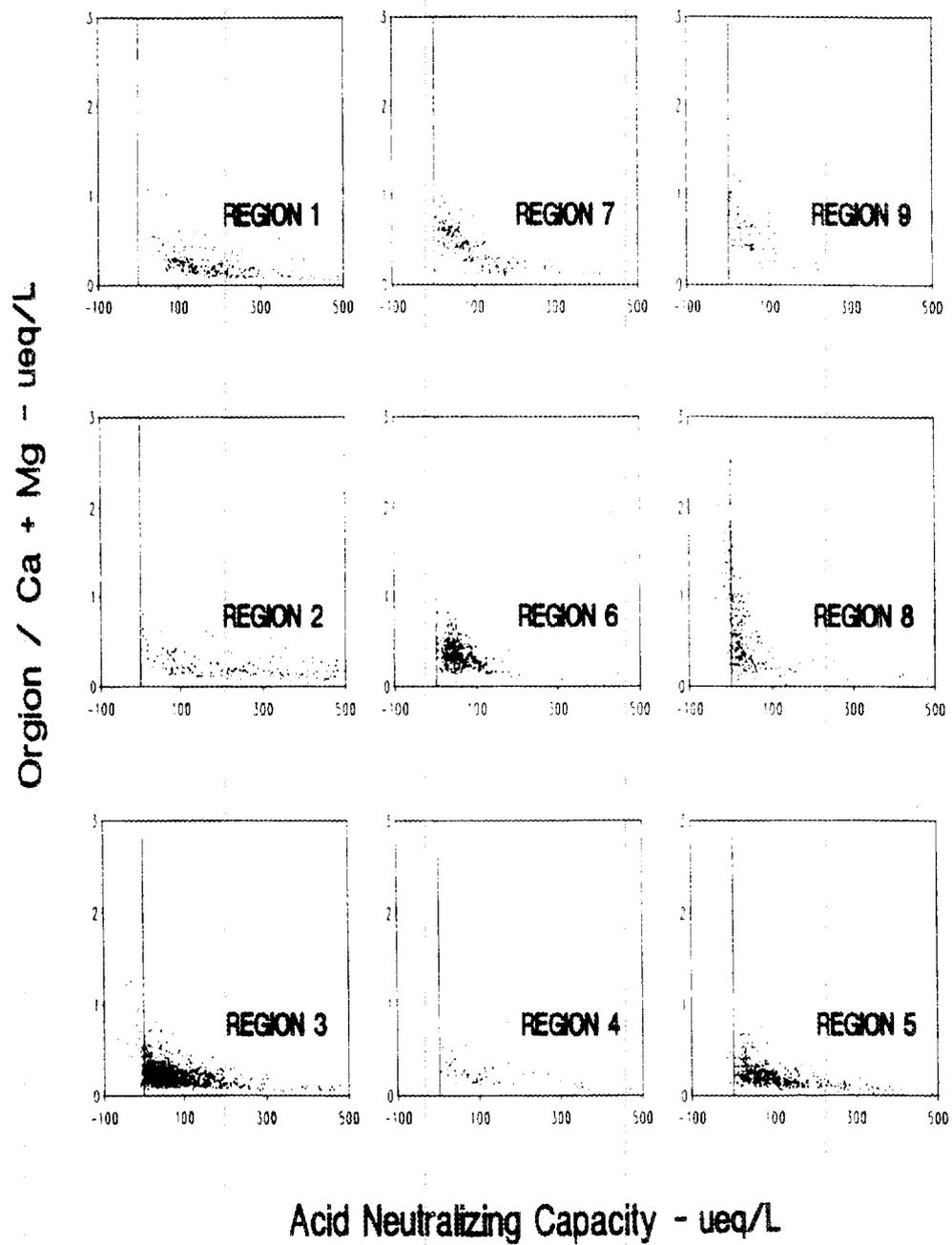


Fig. 25. Organic anion/(orgion)  $[Ca^{2+} + Mg^{2+}]$  vs ANC for Level 3 sampled lakes in southeastern Canada by regions.

of lakes having organic anion:  $[\text{Ca}^{2+} + \text{Mg}^{2+}] > 1$ . Organic anions are present in similar concentrations across southeastern Canada, whereas base cations decline from west to east (Figs. 10 and 11). Thus, the higher values of organic anion:  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  in the North Atlantic and South Atlantic regions compared with those elsewhere in southeastern Canada are the result of a combination of lower concentrations of base cations and similar organic anion concentrations. In southeast Ontario, two low-ANC lakes have an organic anion:  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  ratio greater than one, and two lakes in the West Ontario region have values of this ratio greater than one.

Comparison of Fig. 23 with Fig. 25 reveals that most lakes with  $\text{ANC} \leq 50 \mu\text{eq L}^{-1}$  in South Ontario, North Quebec, southwest Quebec, and Laurentide have low ANC values as a result of  $\text{SO}_4^{2-}$ , not organic anions.  $\text{SO}_4^{2-}$  is also predominant for lakes in the Sudbury/Noranda region (Fig. 24). The North and South Atlantic regions are the only regions in which organic anion is the dominant anion in more low-ANC lakes than is  $\text{SO}_4^{2-}$ .

The influence of atmospheric deposition on lakes having an organic anion to  $[\text{Ca}^{2+} + \text{Mg}^{2+}] > 1$  is not readily apparent. For example, addition of  $\text{SO}_4^{2-}$  to a low-ANC organic-dominated lake may cause acidification without causing a shift in the organic dominance. Low-ANC lakes with organic anion dominant are rarely acidic in Labrador, North Ontario, and West Ontario; these areas receive relatively low levels of atmospheric deposition. In contrast, the South Atlantic region (primarily Nova Scotia) receives moderate levels of atmospheric deposition and has 23 lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  that have organic anion dominant (see the following).

#### 3.5.4 Chemistry of Acidic Lakes

The major ion chemistry of lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  will reveal the combination of anions that contribute to the acidity (Fig. 26). For Level 3 lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  in Sudbury/Noranda,  $\text{SO}_4^{2-}$  is the dominant anion (Table 11). The median organic anion ( $16 \mu\text{eq L}^{-1}$ ; interquartile range:  $5\text{--}26 \mu\text{eq L}^{-1}$ ) in the Sudbury/Noranda acidic lakes is lower than that for the other regions (medians range from 25 to  $51 \mu\text{eq L}^{-1}$ ). Median base cation concentration ( $194 \mu\text{eq L}^{-1}$ ; interquartile range:  $162\text{--}221$ ) in the Sudbury/Noranda acidic lakes is greater than those in the other regions (medians range from 62 to  $109 \mu\text{eq L}^{-1}$ ) (Table 11), perhaps as a result of enhanced base cation production caused by the high current levels of wet (Fig. 2) and dry atmospheric deposition and the higher historic levels associated with emissions from the Sudbury and Noranda smelters. The current wet deposition used in this report does not reflect the historically high levels of wet plus dry  $\text{SO}_4^{2-}$  deposition in Sudbury/Noranda.

Acidic lakes in Southeast Ontario have both lower median  $\text{SO}_4^{2-}$  and base cation concentrations than those in the Sudbury/Noranda region, yet the median ANC values are similar (Table 11). Lakes in the South Atlantic and North Atlantic regions have a relatively low median  $\text{SO}_4^{2-}$  ( $33$  and  $27 \mu\text{eq L}^{-1}$ , respectively); for these lakes organic anion is an important anion (median organic anion concentration:  $51$  and  $25 \mu\text{eq L}^{-1}$ , respectively) (Table 11).

**DOMINANT IONS IN  
ACIDIC LAKES**  
Level 3 lakes with ANC  $\leq 0$

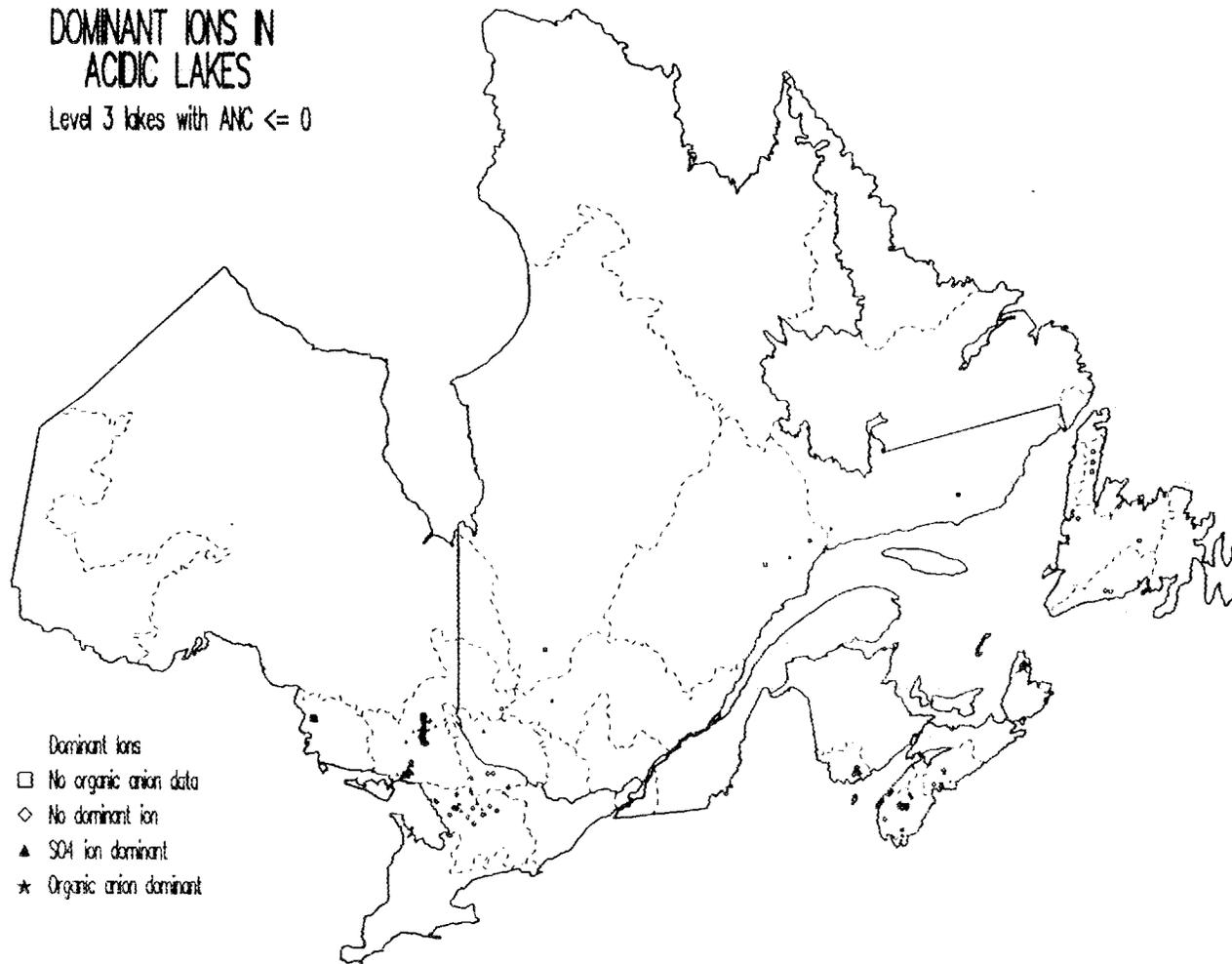


Fig. 26. Map of 230 Level 3 sampled lakes in southeastern Canada with ANC  $\leq 0 \mu\text{eq L}^{-1}$  in which the dominant ion (SO<sub>4</sub><sup>2-</sup>, organic anion, neither, or unknown) is indicated.

Table 11. Chemical characteristics of Level 3 sampled lakes with ANC  $\leq 0$   $\mu\text{eq L}^{-1}$ ; only regions with  $n \geq 10$  are shown. ANC,  $\text{SO}_4^{2-}$ , SBC, and organic anions are expressed as  $\mu\text{eq/L}$  and DOC is expressed as  $\mu\text{mol/L}$

Region	Number of Lakes	pH			ANC			$\text{SO}_4^{2-}$			SBC			Number of Lakes	DOC			Organic Anions		
		Q <sub>1</sub>	Q <sub>2</sub>	Q <sub>3</sub>	Q <sub>1</sub>	Q <sub>2</sub>	Q <sub>3</sub>	Q <sub>1</sub>	Q <sub>2</sub>	Q <sub>3</sub>	Q <sub>1</sub>	Q <sub>2</sub>	Q <sub>3</sub>		Q <sub>1</sub>	Q <sub>2</sub>	Q <sub>3</sub>	Q <sub>1</sub>	Q <sub>2</sub>	Q <sub>3</sub>
3-Southeast Ontario	52	4.8	5.0	5.2	-12	-6	-3	79	92	139	84	109	167	52	258	337	658	23	32	60
8-South Atlantic	57	4.7	4.8	5.0	-16	-8	-2	26	33	44	77	98	120	56	417	587	813	37	51	69
9-North Atlantic	11	4.9	5.2	5.2	-6	-3	-1	23	27	43	54	62	75	5	146	254	442	14	25	39
10-Sudbury/Noranda	108	4.6	4.8	5.0	-27	-18	-9	194	219	243	162	194	221	108	60	171	295	5	16	26

The Sudbury/Noranda region has 106 acidic lakes with median  $\text{SO}_4^{2-} > \text{median} [\text{Ca}^{2+} + \text{Mg}^{2+}]$  (Fig. 27, Table 12). The lake-water chemistry of these lakes (acidic and  $\text{SO}_4^{2-} > [\text{Ca}^{2+} + \text{Mg}^{2+}]$ ) indicates that they have been acidified; the source of the acidity was historically high local atmospheric deposition.

Acidic lakes with median  $\text{SO}_4^{2-}$  greater than median  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  are located in the Muskoka-Haliburton area of southeast Ontario ( $n = 39$ ), the Algoma highlands (near the shore of Lake Superior;  $n = 8$ ). The lake-water chemistry of these lakes also indicates that they have probably experienced some acidification as a result of atmospheric deposition.

Acidic lakes with organic anions as the dominant anions are located in insular Newfoundland and Nova Scotia ( $n = 28$ ) and in Regions 1 through 6 (Fig. 28, Table 12). Organic acids substantially contribute to the acidity of these lakes. In insular Newfoundland and Nova Scotia, 18 lakes have median  $\text{SO}_4^{2-} > [\text{Ca}^{2+} + \text{Mg}^{2+}]$  yet have median organic anions present in concentrations similar to  $\text{SO}_4^{2-}$ . Furthermore, 17 lakes in these two regions have no anion greater than base cation concentrations, and  $\text{SO}_4^{2-}$  and organic anion concentrations are similar. For these 35 lakes, both organic acids and mineral acids contribute to the acid-base chemistry.

An additional 16 acidic lakes in Regions 1 through 6 have  $\text{SO}_4^{2-} > \text{organic anion}$  yet have  $\text{SO}_4^{2-} < [\text{Ca}^{2+} + \text{Mg}^{2+}]$  (Table 12). For these acidic lakes,  $\text{SO}_4^{2-}$  is the predominant anion, yet organic anions are present in relatively high concentrations (Table 11). Thus, the acid-base chemistry of these lakes is the result of a mixture of  $\text{SO}_4^{2-}$  and organic anions.

#### 4. SUMMARY AND CONCLUSIONS

- Lake-water chemistry data from 8506 lakes surveyed in south-eastern Canada were obtained from Environment Canada and the Ontario Ministry of the Environment. Initial quality assurance screening of the lake data resulted in 173 lakes being omitted because of missing key variables (pH, ANC, and  $\text{SO}_4^{2-}$ ), inconsistent pH/ANC, or surface area  $> 2000$  ha. The 8333 remaining lakes have either pH, ANC or  $\text{SO}_4^{2-}$  and are designated Level 1 lakes. Additional lakes were omitted because they were not considered sensitive to acidic deposition (238 lakes had a pH  $> 8$  or an ANC  $> 2000 \mu\text{eq L}^{-1}$ ), had  $\text{Na}^+$  concentrations indicative of road salt (279 lakes), had a charge imbalance (407), had a conductivity imbalance (178 lakes), or were missing any key variables (pH, ANC,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , DOC/color; 3214 lakes). The lakes remaining after this screening ( $n = 4017$ ) had data available for sufficient variables ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , DOC, in addition to pH, ANC, and  $\text{SO}_4^{2-}$ ) (Level 3) to allow more detailed examination of acid-base chemistry on a regional scale.
- The two screening levels (Level 1 and Level 3) had values of ANC, pH, and  $\text{SO}_4^{2-}$  that were similar.

DOMINANT IONS  
IN ACIDIC LAKES  
SUDBURY/NORANDA REGION

Level 3 lakes with ANC  $\leq 0$

- Dominant ions
- No organic anion data
  - ◇ No dominant ion
  - ▲ SO<sub>4</sub> ion dominant
  - ★ Organic anion dominant

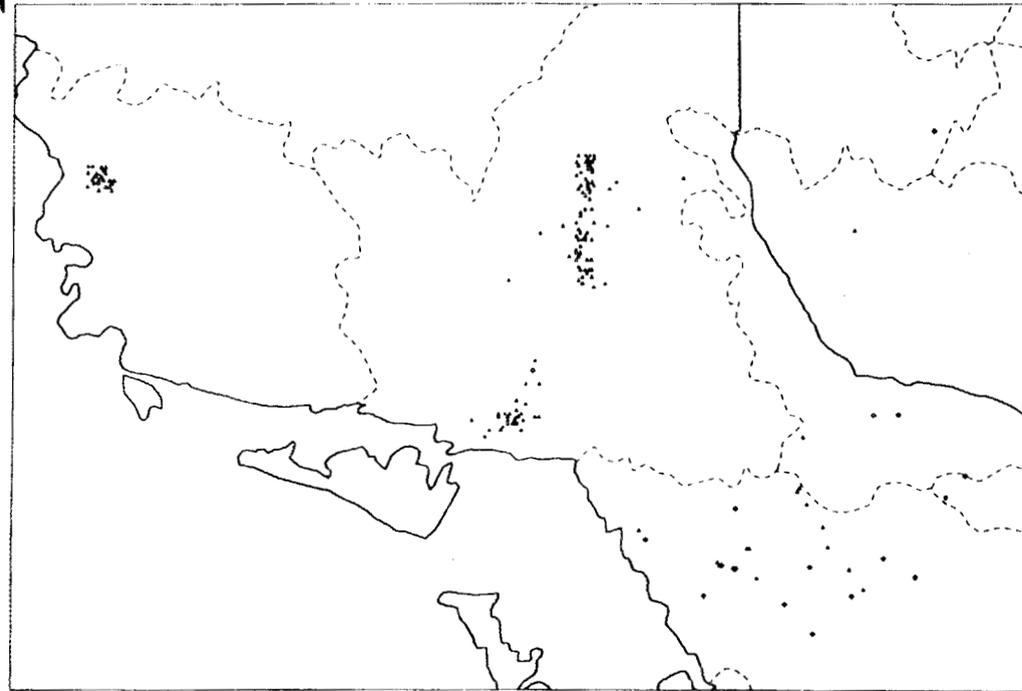


Fig. 27. Map of Level 3 sampled lakes in the southeastern Ontario region with ANC  $\leq 0 \mu\text{eq L}^{-1}$  with the dominant ion (SO<sub>4</sub><sup>2-</sup>, organic anion, neither, or unknown) indicated.

Table 12. Median ANC,  $\text{SO}_4$ , organic anion, and  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  (all expressed as  $\mu\text{eq L}^{-1}$ ) for Level 3 sampled lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$ . Lakes are grouped into three mega-regions: West (Regions 1-6), Sudbury/Noranda (Region 10), and East (Regions 7-9)

		Regions 1-6 (West)	Sudbury/ Noranda (Region 10)	Regions 7-9 (East)
No organic anion data	n			9
	ANC			-5
	$\text{SO}_4^{2-}$			47
	Organic anion			-
	$\text{Ca}^{2+} + \text{Mg}^{2+}$			40
No dominant ion	n	16	2	17
	ANC	-6	-21	-2
	$\text{SO}_4^{2-}$	128	275	30
	Organic anion	66	22	35
	$\text{Ca}^{2+} + \text{Mg}^{2+}$	145	281	58
$\text{SO}_4^{2-}$ ion dominant	n	39	106	18
	ANC	-6	-18	-10
	$\text{SO}_4^{2-}$	87	219	46
	Organic anion	27	17	44
	$\text{Ca}^{2+} + \text{Mg}^{2+}$	73	153	38
Organic anion dominant	n	2		28
	ANC	-38		-10
	$\text{SO}_4^{2-}$	44		27
	Organic anion	126		65
	$\text{Ca}^{2+} + \text{Mg}^{2+}$	101		43

DOMINANT IONS  
IN ACIDIC LAKES

Nova Scotia Region

Level 3 lakes with ANC < 0

- Dominant ions
- No organic anion data
  - ◇ No dominant ion
  - ▲ SO<sub>4</sub> ion dominant
  - ★ Organic anion dominant

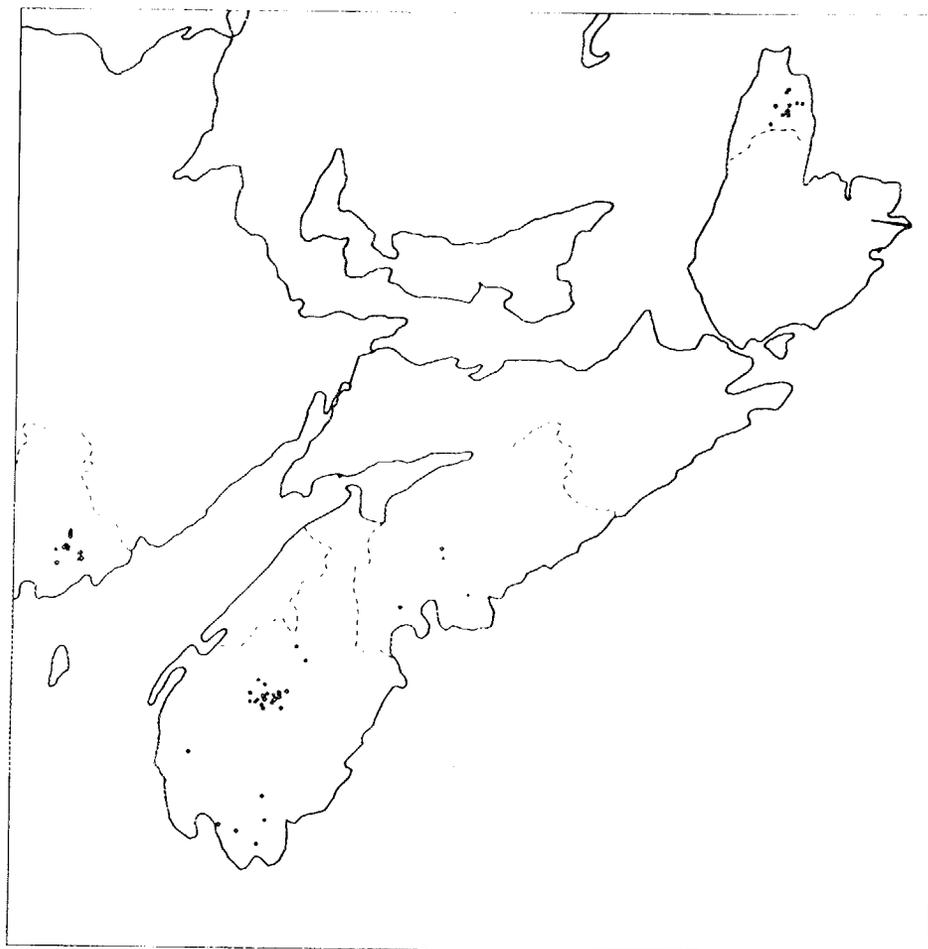
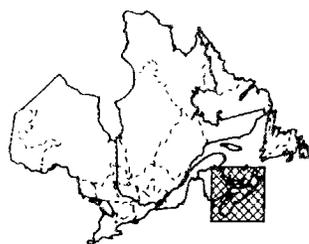


Fig. 28. Map of Level 3 sampled lakes in the south Atlantic region with ANC  $\leq 0 \mu\text{eq L}^{-1}$  in which the dominant ion ( $\text{SO}_4^{2-}$ , organic anion, neither, or unknown) is indicated.

- For the Level 1 lakes in southeastern Canada, 4613 (56% of the 8192 sampled lakes with ANC) have an ANC  $\leq 100 \mu\text{eq L}^{-1}$ . The regions with the largest number of lakes with ANC  $\leq 100 \mu\text{eq L}^{-1}$  are Southeast Ontario (Region 3; 1768 lakes, or 69% of the lakes sampled in this region), the Laurentide (Region 6; 627 lakes, or 84% of the lakes sampled in this region), the South Atlantic (Region 8; 552 lakes, or 72% of the lakes sampled in this region), and Sudbury/Noranda (Region 10; 483, or 71% of the lakes sampled in this region).
- For the Level 1 lakes in southeastern Canada, 3041 (38% of the sampled lakes) have an ANC  $\leq 50 \mu\text{eq L}^{-1}$ . The regions with the largest number of lakes with ANC  $\leq 50 \mu\text{eq L}^{-1}$  are Southeast Ontario (Region 3; 864 lakes, or 41% of the lakes sampled in this region), the South Atlantic (Region 8; 450 lakes, or 59% of the lakes sampled in this region), the Laurentide (Region 6; 445, or 60% of the lakes sampled in this region), and Sudbury/Noranda (Region 10; 402 lakes, or 59% of the lakes sampled in this region).
- Of the Level 1 lakes sampled in southeastern Canada, 381 lakes (5% of the sampled lakes) are acidic (ANC  $\leq 0 \mu\text{eq L}^{-1}$ ). The Sudbury/ Noranda region (Region 10) has the greatest percentage of sampled acidic lakes (24%), and it has 43% of the sampled acidic lakes in all of southeastern Canada. In the South Atlantic region (Region 8), 90 of the 764 sampled lakes (12%) are acidic. In Southeast Ontario (Region 3), 86 (4%) of the sampled lakes are acidic. No acidic lakes were sampled in West Ontario (Region 1), and <1% of the lakes sampled in North Ontario (Region 2), a region characterized by generally high lake-water DOC, are acidic.
- There is a significant positive correlation between lake-water  $\text{SO}_4^{2-}$  and wet  $\text{SO}_4^{2-}$  deposition based on regional median values. This correlation supports the hypothesis that atmospheric deposition is a primary source of  $\text{SO}_4^{2-}$  for many lakes in southeastern Canada; intensive studies of individual watersheds on the Precambrian Shield show that atmospheric deposition is the primary source of  $\text{SO}_4^{2-}$  to these watersheds.
- Within a region, Level 3 lakes with ANC  $\leq 400 \mu\text{eq L}^{-1}$  have higher median base cation content than lakes with ANC  $\leq 50 \mu\text{eq L}^{-1}$ ; median  $\text{SO}_4^{2-}$  and organic anion concentrations are similar between the two ANC strata for each region. The differences in base cation content within regions is probably the result of spatial heterogeneity in base cation production from geologic weathering and cation exchange.
- The absence of a strong relationship between lake-water  $\text{SO}_4^{2-}$  and ANC or  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  and the greater variance in ANC relative to lake-water  $\text{SO}_4^{2-}$  for lakes with ANC  $\leq 400 \mu\text{eq L}^{-1}$  suggests that the lake-water concentrations of ANC and  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$  are controlled

primarily by watershed characteristics, not by atmospheric deposition. For these lakes, any changes caused by deposition are small relative to the natural variation in ANC among lakes.

- For Level 3 lakes in southeastern Canada having  $\text{ANC} \leq 50 \mu\text{eq L}^{-1}$  and  $\text{SO}_4^{2-}$  concentrations similar to or greater than  $[\text{Ca}^{2+} + \text{Mg}^{2+}]$ , both watershed processes and atmospheric deposition control the acid-base chemistry.
- For all regions in southeastern Canada, the pH-ANC data fall below the theoretical pH-ANC (bicarbonate) relationship for ANC concentrations  $> 50 \mu\text{eq L}^{-1}$ . For low-DOC systems, this deviation probably reflects supersaturation with respect to atmospheric  $\text{CO}_2$ . For high-DOC systems, the data are not sufficient to determine which weak acids (i.e.,  $\text{CO}_2$  and organic acids) cause pH to be lower for a given ANC value.
- The solution chemistry of Level 3 lakes in West Ontario (Region 1), North Quebec (Region 7), and North Atlantic (Region 9) appears to consist primarily of base cations and ANC ( $\text{HCO}_3^- +$  organic anions); these three regions receive relatively low levels of atmospheric deposition, have low levels of lake-water  $\text{SO}_4^{2-}$ , and rarely have lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$ . The solution chemistry for the remaining six regions is base cations,  $\text{HCO}_3^-$ , organic anions, and  $\text{SO}_4^{2-}$ .
- Median organic anion concentrations are between 25 and 60  $\mu\text{eq L}^{-1}$  for each of the regions in southeastern Canada, with higher concentrations occurring in North Ontario (Region 2), West Ontario (Region 1), and North Atlantic (Region 9). Organic anions were estimated by using the Oliver et al. (1983) model, which has an uncertainty of up to  $\pm 50\%$ .
- Level 3 lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  and with  $\text{SO}_4^{2-} > [\text{Ca}^{2+} + \text{Mg}^{2+}]$  are primarily located in Sudbury/Noranda (Region 10) (106 out of 230 lakes). These 106 lakes have relatively low organic anion concentrations and have experienced acidification as a result of  $\text{SO}_4^{2-}$  deposition. This region has had historically high levels of atmospheric sulfur deposition from local smelters.
- Level 3 lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  and with  $\text{SO}_4^{2-} > [\text{Ca}^{2+} + \text{Mg}^{2+}]$  are also located in Southeast Ontario (Region 3) and in the Maritime Provinces (Regions 7 through 9). For these acidic lakes in Southeast Ontario, lake-water  $\text{SO}_4^{2-}$  is much greater than organic anion concentrations, suggesting that these lakes have experienced some acidification as a result of atmospheric deposition. For the acidic lakes in Regions 7 through 9 with  $\text{SO}_4^{2-} > [\text{Ca}^{2+} + \text{Mg}^{2+}]$ , organic anions are present at concentrations similar to but less than those of  $\text{SO}_4^{2-}$ , indicating that a combination of mineral acids and organic acids are responsible for the acid-base chemistry.

- Twenty-eight Level 3 lakes with  $\text{ANC} \leq 0 \mu\text{eq L}^{-1}$  and with organic anion  $> [\text{Ca}^{2+} + \text{Mg}^{2+}]$  are located in the South Atlantic region (Region 8; primarily Nova Scotia); these lakes also have organic anions  $> \text{SO}_4^{2-}$ . This combination of anions suggest that the acidity of these 28 lakes is substantially influenced by organic acids.

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APPENDIX A

Lake-Water Chemistry Data For Ontario

Subject: Water Quality

Date: April 7, 1989

Data base Title: Lake-Water Chemistry Data for Ontario

Date Obtained: December 15, 1988

Data Source(s) Contributing to ADDNET: Bernie Neary, Ontario Ministry of Environment

NAPAP Code:

Sponsoring Agency: Department of Energy

Appropriate Reference Citation:

Description: This data set was received from B. P. Neary and is an updated version of ADDNETW.Q62LVLS, which is also a part of ADDNETW.CANLAKES. The original data set was more than 80 columns long, so when it was uploaded to the PDP10, a fortran program separated the data set into three smaller ones with a common ID.

In the first section of the data set:

1. Latitude/longitude hours and minutes were converted to decimal degrees.
2. 232 lakes had missing watershed data. Mapping of these lakes using available latitude and longitude variables produced the watershed data. This information was merged into the data set by ID.
3. In December, 1988, Ontario was divided into three sections based on latitude 45.833 and longitude 85.333. These sections were referred to as NE, SC, and NW.
4. In March, 1989, Ontario was further divided into regions based on tertiary watersheds for the April analysis. These regions were determined as follows:

Region	Watershed
1	2F, 2G, 2HA-2H
2	2E, 2H, 2HF-2HH, 2HJ-2HM, 2K-2M, 2O
3	2D, 2J, 2JC-2JE
4	2BD-2BF, 2C
5	4G, 4H, 4J-4N
6	2A, 2B, 2BA-2BC
7	5P-5R
8	4A-4F

- In August, 1989, Ontario and the other provinces were divided into nine regions based on a grouping of tertiary watersheds provided by Dean Jeffries, Environment Canada. Ontario contains six regions.
- Variables were labelled as follows:

```

NAME = 'LAKE NAME'
ID = 'SEQUENTIAL NUMBERS ASSIGNED BY ORNL'
LAT_DEC = 'LATITUDE, DECIMAL DEGREES'
LON_DEC = 'LONGITUDE, DECIMAL DEGREES'
PROVINCE = 'CANADIAN PROVINCE'
SECTION = 'SECTION OF ONTARIO'
REGION = 'REGION'
MNR = 'LAKE LOCALE - DIST. MIN. NAT. RES.'
```

In the second section of the data set:

- Missing values were set to '.', and obviously incorrect values (i.e. day > 31 or pH > 14) were also set to missing (.).
- These variables were calculated as follows:

```

ANC = TIPALK * 20
DATE = MDY(MONTH, DAY, YEAR)
      FORMAT DATE DATE7.
H_ION = 10**(-PH) * 10**6
HCO3 = ANC + H_ION
PK = 0.96 + 0.9 * PH - 0.039 * PH**2
K1 = 10**(-PK)
RATIO = LK_AREA / SHEDAREA
```

- Variables were labelled as follows:

```

H_ION = '10**(-PH) * 10**6'
HCO3 = 'ANC + H_ION'
PK = '0.96 + 0.9 * PH - 0.039 * PH**2'
K1 = '10**(-PK)'
LK_AREA = 'LAKE AREA (HECTARES)'
SHEDAREA = 'WATERSHED AREA (HECTARES)'
RATIO = 'LK_AREA/SHEDAREA'
TWNShP = 'LAKE LOCATION'
```

DATE = 'DATE SAMPLE TAKEN'  
 MONTH = 'MONTH SAMPLE TAKEN'  
 DAY = 'DAY SAMPLE TAKEN'  
 YEAR = 'YEAR SAMPLE TAKEN'  
 PH = 'SAMPLE PH'  
 TIPALK = 'TOT INFLEC PT ALK (GRAN TITRA.,MG/L)'  
 CONDOC = 'CONDUCTIVITY (MICRO SIEMENS)'  
 AP\_COLOR = 'APPAR COLOR, UNFILT SMPL (HAZEN UNITS)'  
 TRUCOLOR = 'COLOR OF FILTERED SAMPLE (HAZEN UNITS)'  
 ANC = 'ACID NEUTRALIZING CAPACITY'

In the third section of the data set:

1. Missing values were set to '.'.
2. Negative chloride and total aluminum values were corrected with the following equations:

$$\begin{aligned}
 \text{ALTL} &= (-0.5) * (-\text{ALTL}) \\
 \text{CL\_MEAS} &= (-0.5) * (-\text{CL\_MEAS})
 \end{aligned}$$

3. Units changed from MG/L to UEQ/L .

$$\begin{aligned}
 \text{DOC1\_UM} &= \text{DOC} * 83.3 \\
 \text{CA} &= 49.9 * \text{CA} \\
 \text{MG} &= 82.26 * \text{MG} \\
 \text{NA} &= 43.5 * \text{NA} \\
 \text{K} &= 25.57 * \text{K} \\
 \text{SO4} &= 20.82 * \text{SO4} \\
 \text{CL\_MEAS} &= 28.21 * \text{CL\_MEAS}
 \end{aligned}$$

4. Variables calculated:

$$\begin{aligned}
 \text{CA\_MG} &= \text{SUM} (\text{CA}, \text{MG}) \\
 \text{SBC} &= \text{NA} + \text{K} + \text{CA} + \text{MG}
 \end{aligned}$$

5. Variables were labelled as follows:

DOC = 'DISSOLVED ORGANIC CARBON (MG/L)'  
 CA = 'CALCIUM (UEQ/L)'  
 MG = 'MAGNESIUM (UEQ/L)'  
 NA = 'SODIUM (UEQ/L)'  
 K = 'POTASSIUM (UEQ/L)'  
 SO4 = 'SULFATE (UEQ/L)'  
 CL\_MEAS = 'MEASURED CHLORIDE (UEQ/L)'  
 ALTL = 'TOTAL ALUMINUM (MG/L)'  
 MN = 'MANGANESE (MG/L)'  
 FE = 'IRON (MG/L)'  
 CA\_MG = 'CA + MG'  
 SBC = 'SOLUB BASE CATIONS=NA + K + CA + MG'  
 DOC1\_UM = 'MEAS. DISSOLVED ORGANIC CARBON (UM)'  
 DATACAT = 'DATA CATEGORY'

These data sets were sorted, and then merged by ID. At this point, regression equations to calculate DOC from apparent and true color and chloride from sodium were inserted into the data set. These two variables are important in the statistical analysis and, therefore, in the QA/QC assessment of this data set. Of the 5366 observations, there were 3100 missing DOC values (most of which were in the NW section) and 4832 missing Cl values. Because of this deficiency, the following equations were used to supplement measured values:

$$\begin{aligned} \text{DOC2\_UM} &= (2.2115 + 0.0927 * \text{AP\_COLOR}) * 83.3 \\ \text{DOC3\_UM} &= (3.1871 + 0.1163 * \text{TRUCOLOR}) * 83.3 \\ \text{CL} &= (0.32 * \text{NA}) - 1.8962 \end{aligned}$$

DOC\_UM was set to equal DOC1\_UM. If DOC1\_UM was missing, values calculated from AP\_COLOR were inserted and then any missing values were set to equal those calculated from TRUCOLOR. Use of these equations increased the number of observations available for QA/QC by 808 for DOC (474 of which were in the NW section) and by 2491 for Cl (an increase of 989 for NE, 543 for NW, and 959 for SC).

Although the R-square values were low (.6 and .2 for DOC and Cl, respectively), other considerations were taken into account. For both DOC and Cl, calculated values were within the range of measured values. The only exception to this was for Cl in the SC section, where there were only 19 of the 534 measured chloride values compared with 978 calculated values. Ultimately the range for SC section was more in line with ranges of the other two sections.

The following variables were calculated at this point:

$$\begin{aligned} \text{CT1} &= \text{DOC} * 10 \\ \text{CT2} &= ((2.2115 + (0.0927 * \text{AP\_COLOR})) * 10) \\ \text{CT3} &= ((3.1871 + (0.1163 * \text{TRUCOLOR})) * 10) \\ \text{ORGION1} &= \text{K1} * \text{CT1} / (\text{K1} + (10 ** (-\text{PH}))) \\ \text{ORGION2} &= \text{K1} * \text{CT2} / (\text{K1} + (10 ** (-\text{PH}))) \\ \text{ORGION3} &= \text{K1} * \text{CT3} / (\text{K1} + (10 ** (-\text{PH}))) \\ \text{ANIONS} &= \text{SUM} (\text{HCO3}, \text{CL}, \text{SO4}, \text{ORGION}) \\ \text{CATIONS} &= \text{SUM} (\text{CA}, \text{MG}, \text{K}, \text{NA}, \text{H\_ION}) \\ \text{SO4\_CAT} &= \text{SO4} / \text{CATIONS} \\ \text{ANC\_SO4} &= \text{SUM} (\text{ANC}, \text{SO4}) \\ \text{SO4\_CAMG} &= \text{SO4} / \text{SUM} (\text{CA}, \text{MG}) \\ \text{ORG\_CAMG} &= \text{ORGION} / \text{SUM} (\text{CA}, \text{MG}) \\ \text{COND\_CAL} &= \text{SUM} (\text{CA} * 59.47, \text{MG} * 53.0, \text{K} * 73.48, \text{NA} * 50.08, \\ &\quad \text{H\_ION} * 349.65, \text{HCO3} * 44.5, \text{SO4} * 80.0, \text{CL} * 76.31) \\ \text{COND\_CAL} &= \text{COND\_CAL} / 1000 \\ \text{ORG\_CAT} &= \text{ORGION} / \text{CATIONS} \\ \text{CAT\_AN} &= \text{CATIONS} / \text{ANIONS} \end{aligned}$$

Orgion was calculated to estimate the organic ion concentration using Oliver's method from the National Surface Water Survey: Eastern Lake Survey-Phase I, Data Base Dictionary, P. Kanciruk, et al., ORNL/TM-10153. Measured and calculated values were combined as those for DOC\_UM and variable was called ORGION.

Criteria to determine anion/cation balance and conductance balance were drawn from USEPA Corvallis, Working Protocol for Long-term Monitoring Program, May 1985. Calculations necessary are:

$$\begin{aligned} \text{DIFF\_A\_C} &= \text{ANIONS} - \text{CATIONS} \\ \text{ION\_STRG} &= \text{CATIONS} + \text{ANIONS} \\ \text{DIFF\_ION} &= ( \text{ABS}(\text{DIFF\_A\_C}) / \text{ION\_STRG} ) * 100 \\ \text{DIFFCOND} &= ( ( \text{ABS}(\text{COND\_CAL} - \text{CONDOC}) ) / \text{CONDOC} ) * 100 \end{aligned}$$

Chemical analysis criteria:

1. Cation/Anion Balance

If	Total Ion Strength ( $\mu\text{eq L}^{-1}$ )	and	Percentage Ion Difference	then	Ion Class
	< 50		< $\pm$ 60		50
	$\geq$ 50 < 100		< $\pm$ 30		75
	$\geq$ 100		< $\pm$ 15		100
				else	0

2. Conductance Balance

If	Measured Conductance ( $\mu\text{S cm}^{-1}$ )	and	Percentage Conductance Difference	then	Conductivity Class
	< 5		< 50		5
	$\geq$ 5 < 30		< 30		15
	$\geq$ 30		< 20		30
				else	0

In the first data set of Ontario lakes, a variable called 'SOURCE' reflected the study under which the sample was taken. To have that information available in this data set, lakes were matched by name, latitude, longitude, township, and SOURCE was added to 4748 observations.

Each observation was then tested and assigned to a 'LEVEL', depending on whether it passed or failed specific, prioritized criteria. If an observation failed to meet a criteria a 'FLAG' was assigned to describe the reason. Only Level 0 lakes had incorrect or enough missing information to disqualify them from use in analysis. Levels are described as follows:

LEVEL	FLAG	No. of OBS.	DESCRIPTION												
0		159	If 'FLAGS' are 1 to 5 then observations fell into this level.												
	1	0	LOCATION - Latitude and/or lngitude incorrect												
	2	146	AREA - Lake area > 2000												
	3	2	ALLMISS - All primary variables missing (pH, ANC, SO4)												
	4	11	BADPHANC - Plots of pH vs ANC showed these outliers, ID's are: <div style="margin-left: 100px;"> <table border="0"> <tr> <td>43</td> <td>1207</td> <td>2938</td> </tr> <tr> <td>96</td> <td>1538</td> <td>3333</td> </tr> <tr> <td>229</td> <td>1669</td> <td>3865</td> </tr> <tr> <td>896</td> <td>1713</td> <td></td> </tr> </table> </div>	43	1207	2938	96	1538	3333	229	1669	3865	896	1713	
43	1207	2938													
96	1538	3333													
229	1669	3865													
896	1713														
	5	0	BADVAR - Inconsistant analytical results												
	1	2335	If 'FLAGS' are 6 to 8 then observations fell into this level.												
	6	2117	KEYMISS - At least one key variable is missing (pH, ANC, SO4, Ca, Mg)												
	7	141	HIANC - ANC > 2000												
	8	77	HIPH - pH > 8.0												
2		621	If 'FLAGS' are 9 to 12 then observations fell into this level.												
	9	422	OTHERVAR - At least one variable missing that is needed for charge balance check (pH, ANC, SO4, Na, DOC, K conductivity)												
	10	111	HINA - Na > 87												
	11	71	CHGIMBAL - Cation/anion imbalance												
	12	17	CONIMBAL - Conductivity imbalance												
3	0	2251	GOODDATA - Data passed all criteria and fell into this level												

Lakes located near Sudbury Smelter in southern Ontario were flagged to represent distance from the plant. There were about 475 lakes within a radius of 100 kilometers that are designated as follows:

Sudbury Flag	Description
0	Outside of the 100 km radius
1	0-25 km radius
2	25-50 km radius
3	50-75 km radius
4	75-100 km radius

**Data Uses:** Comparison of Canadian lakes in Ontario to determine regional patterns of acidification or differences in ionic balance caused by deposition and terrain sensitivity.

**Data Importance:** This is a comprehensive , QA/QC data set of water quality status of lakes in Ontario, Canada allowing the determination of possible effects of acid deposition on lake water chemistry.

**Data Type:** Derived

**Number of Records:** 9783

**Geographic Coverage:** Ontario, Canada

**Spacial Resolution:** Point, expressed by latitude-longitude coordinate

**Period of Record:** 1978-1987

**Temporal Resolution:** Point

**References or Supporting Documents:**

Neary, B. P., and P. J. Dillon. Effects of Sulfur Deposition on Lake-water Chemistry in Ontario, Canada. *Nature* 333:340-343 (1988).

Kanciruk, P., J. M. Eilers, R. J. McCord, D. H. Landers, D. F. Brakke, and R. A. Linthurst. Characteristics of Lakes in the Eastern United States. Volume III. Data Compendium of Site Characteristics and Chemical Variables. EPA/600-4-86/007c, U. S. Environmental Protection Agency, Washington, DC, 1986, 439 pp.

## Lake-Water Chemistry Data for Southeastern Canada

Subject: Water Quality

Date: August 25, 1989

Data base Title: Lake-Water Chemistry Data for Southeastern Canada

Date Obtained: June 27, 1989

Data Source(s) Contributing to ADDNET: Dean Jeffries, R. L. Thomas  
Environment Canada

NAPAP Code:

Sponsoring Agency: Department of Energy

Appropriate Reference Citation:

Description: This data was received from R. L. Thomas and is an updated version of ADDNETW.Q61, which is also a part of ADDNETW.CANLAKES. Data were received for 3140 lakes in Quebec, New Brunswick, Nova Scotia, Prince Edward Island, and Newfoundland. Data were also sent for Ontario, but we were informed by Jeffries that those data should be the same as the data we received from Neary, and we opted to use the more complete Neary data for Ontario.

Differences between the Jeffries and Neary data set included: medians and number of observations (n=1 to >100) for each lake; both raw and sea salt corrected data provided for NF, NS, PEI, NB, and part of Quebec; NO<sub>3</sub> and NH<sub>4</sub> provided for some lakes, and no lake area, watershed area, or lake names included.

The data set was more than 80 columns long so when it was uploaded to the PDP10 a fortran program separated the data set into four smaller files with a common ID. Each file was processed and then combined by ID. The coordinates for each lake were sent in separate files and needed to be merged with the chemistry using lake ID.

In the first section of the data set:

6. Variables were labelled as follows:

SOURCE	=	'DATA SOURCE'
WTRSHD	=	'WATERSHED (TERTIARY LEVEL)'
SUB_SHD	=	'WATERSHED SUBDIVISION'
ID	=	'SEQUENTIAL NUMBERS ASSIGNED BY ORNL'
LAT_DEC	=	'LATITUDE, DECIMAL DEGREES'
LON_DEC	=	'LONGITUDE, DECIMAL DEGREES'
PROVINCE	=	'CANADIAN PROVINCE'
REGION	=	'REGION'

In the second section of the data set:

1. Missing values were set to '.' and obviously incorrect values (i.e. day > 31 or pH > 14) were also set to missing (.).
2. These variables were calculated as follows:

$$\begin{aligned} \text{H\_ION} &= 10^{**(-\text{PH})} * 10^{**6} \\ \text{HCO3} &= \text{ANC} + \text{H\_ION} \\ \text{PK} &= 0.96 + 0.9 * \text{PH} - 0.039 * \text{PH}^{**2} \\ \text{K1} &= 10^{**(-\text{PK})} \end{aligned}$$

3. Variables were labelled as follows:

$$\begin{aligned} \text{H\_ION} &= '10^{**(-\text{PH})} * 10^{**6}' \\ \text{HCO3} &= '\text{ANC} + \text{H\_ION}' \\ \text{PK} &= '0.96 + 0.9 * \text{PH} - 0.039 * \text{PH}^{**2}' \\ \text{K1} &= '10^{**(-\text{PK})}' \\ \text{PH} &= '\text{SAMPLE PH}' \\ \text{CONDUCT} &= '\text{CONDUCTIVITY (MICRO SIEMENS)}' \\ \text{ANC} &= '\text{ACID NEUTRALIZING CAPACITY}' \end{aligned}$$

In the third section of the data set:

1. Missing values were set to '.'.
2. NO<sub>3</sub>-NO<sub>2</sub> and NH<sub>3</sub> were expressed as the dissolved nitrate/nitrite and dissolved ammonia; estimates of NO<sub>3</sub> and NH<sub>4</sub> were made using the following equations:

$$\begin{aligned} \text{NO3} &= \text{NO2\_3} * 4.4266 \\ \text{NH4} &= \text{NH3} * 1.2878 \end{aligned}$$

3. Units changed from MG/L to UEQ/L:

$$\begin{aligned} \text{DOC\_UM} &= \text{DOC} * 83.3 \\ \text{CA} &= 49.9 * \text{CA} \\ \text{MG} &= 82.26 * \text{MG} \\ \text{NA} &= 43.5 * \text{NA} \\ \text{K} &= 25.57 * \text{K} \\ \text{SO4} &= 20.82 * \text{SO4} \\ \text{ANC} &= 20.0 * \text{ANC} \\ \text{CL} &= 28.21 * \text{CL} \\ \text{NO3} &= 16.13 * \text{NO3} \\ \text{NH4} &= 55.44 * \text{NO3} \end{aligned}$$

4. Variables calculated:

$$\begin{aligned} \text{CA\_MG} &= \text{SUM} (\text{CA}, \text{MG}) \\ \text{SBC} &= \text{NA} + \text{K} + \text{CA} + \text{MG} \end{aligned}$$

5. Variables were labelled as follows:

```

DOC   = 'DISSOLVED ORGANIC CARBON (MG/L)'
CA    = 'CALCIUM (UEQ/L)'
MG    = 'MAGNESIUM (UEQ/L)'
NA    = 'SODIUM (UEQ/L)'
K     = 'POTASSIUM (UEQ/L)'
SO4   = 'SULFATE (UEQ/L)'
CL_MEAS = 'MEASURED CHLORIDE (UEQ/L)'
ALTL  = 'TOTAL ALUMINUM (MG/L)'
MN    = 'MANGANESE (MG/L)'
FE    = 'IRON (MG/L)'
CA_MG = 'CA + MG'
SBC   = 'SOLUB BASE CATIONS=NA + K + CA + MG'
DOC_UM = 'MEAS. DISSOLVED ORGANIC CARBON (UM)'
DATACAT = 'DATA CATEGORY'

```

The following variables were calculated at this point:

```

CT      = DOC*10
ORGION  = K1*CT / (K1+(10**(-PH)))
ANIONS  = SUM (HCO3, CL, SO4, ORGION, NO3)
CATIONS = SUM (CA, MG, K, NA, H_ION, NH4)
SO4_CAT = SO4 / CATIONS
ANC_SO4 = SUM(ANC,SO4)
SO4_CAMG = SO4 / SUM(CA,MG)
ORG_CAMG = ORGION / SUM(CA,MG)
COND_CAL = SUM ( CA*59.47, MG*53.0, K*73.48, NA*50.08,
                H_ION*349.65, SO4*80.0, CL*76.31)
COND_CAL = COND_CAL / 1000
ORG_CAT  = ORGION / CATIONS
CAT_AN   = CATIONS / ANIONS
C_ANC    = SBC - SO4 - CL - NO3

```

Orgion was calculated to estimate the organic ion concentration using Oliver's method from the National Surface Water Survey: Eastern Lake Survey-Phase I, Data Base Dictionary, P. Kanciruk, et al., ORNL/TM-10153.

Criteria to determine anion/cation balance and conductance balance were drawn from USEPA Corvallis, Working Protocol for Long-term Monitoring Program, May 1985. These calculations were made before se salt corrections were performed. Equations are:

```

DIFF_A_C = ANIONS - CATIONS
ION_STRG = CATIONS + ANIONS
DIFF_ION = ( ABS(DIFF_A_C) / ION_STRG ) * 100
DIFFCOND = ( (ABS(COND_CAL - CONDOC)) / CONDOC ) * 100

```

## 1. Cation/Anion Balance

If	Total Ion Strength ( $\mu\text{eq L}^{-1}$ )	and	Percentage Ion Difference	then	Ion Class
	< 50		< $\pm 60$		50
	$\geq 50 < 100$		< $\pm 30$		75
	$\geq 100$		< $\pm 15$		100
				else	0

## 2. Conductance Balance

If	Measured Conductance ( $\mu\text{S cm}^{-1}$ )	and	Percentage Conductance Difference	then	Conductivity Class
	< 5		< 50		5
	$\geq 5 < 30$		< 30		15
	$\geq 30$		< 20		30
				else	0

Sea-salt corrections were made using the following equations (any negative calculated values were set to zero):

$$\begin{aligned} \text{NA} &= \text{NA} - (\text{CL} * 0.556428) \\ \text{K} &= \text{K} - (\text{CL} * 0.020021) \\ \text{CA} &= \text{CA} - (\text{CL} * 0.021075) \\ \text{MG} &= \text{MG} - (\text{CL} * 0.067018) \\ \text{SO4} &= \text{SO4} - (\text{CL} * 0.139568) \end{aligned}$$

Corrected latitudes and longitudes were entered into the data set as were 22 regions based on tertiary watersheds. These regions were labelled REGION22. Later, ten "mega-regions" were produced from tertiary watershed aggregates.

Each observation was then tested and assigned to a 'LEVEL', depending on whether it passed or failed specific, prioritized criteria. If an observation failed to meet a criteria a 'FLAG' was assigned to describe the reason. Only Level 0 lakes had incorrect or enough missing information to disqualify them from use in analysis. Levels are described as follows:

LEVEL	FLAG	No. of OBS.	DESCRIPTION
0		14	If 'FLAGS' are 1 to 5 then observations fell into this level.
	1	0	LOCATION - Latitude and/or longitude incorrect
	2	NA	AREA - Lake area > 2000 (not applicable in this data set)
3	5		ALLMISS - All primary variables missing (pH, ANC, SO4)
4	2		BADPHANC - Plots of pH vs ANC showed these outliers, ID's are: 7825            7975
5	7		BADVAR - Inconsistent analytical results. ID's are: 6851            8497            8410            6311 6852            8499            8494
1		651	If 'FLAGS' are 6 to 8 then observations fell into this level.
	6	591	KEYMISS - At least one key variable is missing (pH, ANC, SO4, Ca, Mg)
	7	2	HIANC - ANC > 2000
	8	18	HIPH - pH > 8.0
2	749		If 'FLAGS' are 9 to 12 then observations fell into this level.
9	84		OTHERVAR - At least one variable missing that is needed for charge balance check (pH, ANC, SO4, Na, Cl, K, conductivity)
	10	168	HINA - Na > 87
	11	336	CHGIMBAL - Cation/Anion imbalance
	12	161	CONIMBAL - Conductivity imbalance
3	0	1746	GOODDATA - Data passed all criteria and fell into this level

**Data Uses:** Comparison of Canadian lakes to determine regional patterns of acidification or differences in ionic balance caused by deposition and terrain sensitivity.

**Data Importance:** This is a comprehensive , QA/QC data set of water quality status of lakes, Canada allowing the determination of possible effects of acid deposition on lake water chemistry.

**Data Type:** Derived

**Number of Records:**

**Geographic Coverage:** Ontario, Quebec, Newbrunswick, Nova Scotia, Newfoundland, Prince Edward Island; Canada

**Spacial Resolution:** Point, expressed by latitude-longitude coordinate

**Period of Record:** 1980-1988

**Temporal Resolution:** Point

**References or Supporting Documents:**

Neary, B. P., and P. J. Dillon. Effects of Sulfur Deposition on Lake-water Chemistry in Ontario, Canada. Nature 333:340-343. (1988).

Kanciruk, P., J. M. Eilers, R. J. McCord, D. H. Landers, D. F. Brakke, and R. A. Linthurst. Characteristics of Lakes in the Eastern United States. Volume III. Data Compendium of Site Characteristics and Chemical Variables. EPA/600-4-86/007c, U. S. Environmental Protection Agency, Washington, DC, 1986, 439 pp.

Canadian Tertiary Watershed Aggregates

Subject: Regional Stratification

Date: August 25, 1989

Database Title: Canadian Tertiary Watershed Aggregates

Date Obtained: July 25, 1989

Data Source(s) Contributing to ADDNET: Nichole Cartrand, Environment Canada (tertiary watershed boundaries) Dean Jeffries, Environment Canada (aggregation)

NAPAP Code:

Sponsoring Agency: Department of Energy

Appropriate Reference Citation:

Description: Digital data defining the provinces, tertiary watersheds, and ecodistricts for Ontario, Quebec, and the Atlantic provinces were obtained from Environment Canada as SPANS files. The files were exported to ARC/INFO to allow additional analysis and cartographic display.

Lake chemistry data were provided for 8504 lakes falling within 304 tertiary watersheds across eastern Canada. To obtain geographical units amenable to presentation of spatial trends, tertiary watershed units were grouped into 22 aggregates. The intent was to obtain groupings of lakes that are reasonably homogeneous with respect to geology, climate, and atmospheric deposition. As a general indicator of the influence of geology on water chemistry, specific conductance data for the lakes were segregated into classes (<25, 25-49, 50-99, >100 uS), and the classes were then plotted on maps. The general regional groupings suggested by these maps when combined with knowledge of geology, terrain characteristics, and deposition (including consideration of both long-range and local sources) led to specification of 22 tertiary watershed aggregates across eastern Canada. The smallest possible subunit making up an aggregate is a tertiary watershed, and in some cases, tertiary watersheds cross major geological boundaries. Certain aggregates (7, 15, 17) are in fact composed of two sections that are separated by another aggregate chosen to reflect a local emission source (e.g., Sudbury-Noranda, Aggregate 19), localized geochemical influences (e.g., Southeastern Ontario and the Ottawa River Valley, Aggregate 16), or differing geomorphology (Aggregate 7 in Newfoundland). The Canadians believe that the combination of objective and subjective criteria to define the aggregates provides reasonable geographic subdivisions. In Nova

Scotia, New Brunswick, and Newfoundland (an area having relatively low gradients in deposition), purely objective cluster analysis gives nearly identical data subgroups. (Description provided in letter from Jeffries, dated July 10, 1989.)

The digital boundaries for the 22 watershed aggregates were obtained by aggregating the watershed units using the SPANS system and data provided by Environment Canada. The boundaries were transferred to ARC/INFO for mapping.

The 22 units were reduced to 10 for analysis and display for the SOS/T report. Our regions consist of seven regions (1-6 and 10) in Ontario and Quebec that correspond directly to 7 of the 22 watershed aggregates defined by Environment Canada. We grouped 12 watershed aggregates covering the Atlantic provinces into three contiguous regions (7, 8, and 9), based on the smaller number of sampled lakes and lower levels and deposition gradients. Avalon, Newfoundland (Aggregate 5, influenced by urban area), Belle Island, Newfoundland (Aggregate 9, dominated by limestone geology), and South Ontario (Aggregate 18, dominated by limestone geology without any sampled lakes) were dropped from the analysis.

**Data Uses:** Regional stratification was used in comparison of Canadian lakes to determine regional patterns of acidification or differences in ionic balance caused by deposition and terrain sensitivity.

**Data Importance:** This is a physiographic based stratification.

**Data Type:** Digital boundaries and tabular.

**Number of Records:** 415 tertiary watersheds in 6 provinces, 22 aggregated tertiary watersheds, 10 regions.

**Geographic Coverage:** Ontario, Quebec, New Brunswick, Nova Scotia, Newfoundland, Prince Edward Island; Canada

**Spacial Resolution:** Polygons

**Period of Record:** 1989

**Temporal Resolution:** Polygons

**References or Supporting Documents:** Personal Communication from Jeffries, D. S. Environment Canada. July 1989.

Atmospheric Deposition for Southeastern Canada

**Subject:** Atmospheric Deposition

**Date:** April 5, 1990

**Database Title:** 1982-86 Average Excess  $\text{SO}_4^{2-}$  Deposition for approximate 0.5° grid cells

**Date Obtained:** October 4, 1989

**Data Source(s) Contributing to ADDNET:**

Robert Vet, Quality Assurance Data Management Section, Atmospheric Environment Service, Environment Canada, 4905 Dufferin Street, Downsview, Ontario, Canada, Phone: (416) 739-4853

**NAPAP Code:**

**Sponsoring Agency:** Department of Energy

**Appropriate Reference Citation:**

**Description:** Five-year average (1982-86) wet deposition ( $\text{g/m}^2\text{m}$ ) and precipitation concentration ( $\text{mg/L}$ ) estimates of excess sulfate for approximate 0.5° grid cells for an area covering eastern Canada, south of 52°N, were provided from the NatChem by Environment Canada. Data are provided for cells north of 52° but are subject to uncertainty because of the lack of monitoring sites. The data were generated by universal kriging from point data (>100 POINTS) from various monitoring networks (APIOS-C, APIOS-D, APN, CANSAP, CAPMON, EPS, MAP3S/PCN, MNPC, NADP/NTN, NB1WP, NS1WP, QB1WP, AND UAPSP). Grid files were generated for each year based on available data and the five values averaged to produce the mean estimates at the center of grid each cell. These data are consistent with similar deposition data being used for aquatic studies being conducted for the NAPAP SOS/T report from the ADS system.

Dry deposition in Canada was assumed to represent a uniform 15% of the total (wet plus dry) deposition. Dean Jeffries adjusted the data accordingly but increased the 15% to 50% in Sudbury, Ontario and to 35% in the southern part of the Avalon Peninsula, Newfoundland, and in the Halifax area, Nova Scotia.

**Data Uses:** These data were used to estimate deposition for individual lakes and to define regional patterns.

**Data Importance:** Deposition is essential to explaining variation in lake-water chemistry.

**Data Type:** Grid cells extrapolated from point data

**Number of Records:** 4461

**Geographic Coverage:** Ontario, Quebec, New Brunswick, Nova Scotia, Newfoundland, Prince Edward Island; Canada

**Spacial Resolution:** Approximately 0.5°

**Period of Record:** 1982-86

**Temporal Resolution:** 5-year annual average

**References or Supporting Documents:** Personal Communication from Vet, R. Environment Canada. October 4, 1989.



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