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A Survey of Dilute Lakes on the South Coast of Insular Newfoundland in Relation to Natural and Anthropogenic Acidification

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October 1989

Canadian Technical Report of Fisheries and Aquatic Sciences No. 1711



Canadian Technical Report of Fisheries and Aquatic Sciences

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A SURVEY OF DILUTE LAKES ON THE SOUTH COAST

OF INSULAR NEWFOUNDLAND IN RELATION

TO NATURAL AND ANTHROPOGENIC ACIDIFICATION

by

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@Minister of Supply and Services Canada 1989
Cat. No. Fs 97-6/1711E ISSN 0706-6457

Correct citation for this publication:

Scruton, D. A., and D. H. Taylor. 1989. A survey of dilute lakes on the south coast of insular Newfoundland in relation to natural and anthropogenic acidification. Can. Tech. Rep. Fish. Aquat. Sci. 1711: v + 58 p.

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ABSTRACT

Scruton, D. A., and D. H. Taylor. 1989. A survey of dilute lakes on the south coast of insular Newfoundland in relation to natural and anthropogenic acidification. Can. Tech. Rep. Fish. Aquat. Sci. 1711: v + 58 p.

In July 1983, a synoptic survey of the water chemistry of 90 lakes on the south coast of insular Newfoundland was undertaken to address sensitivity to and effects from the Long Range Transport of Air Pollutants (LRTAP). Study lakes were characterized by very low alkalinities (all $\leq 200~\mu eq~L^{-1}$, x = 23.7) and acidic pH values (mostly less than pH 6.50, x = 5.76). Lakes ranged from clear to high coloured with 51 lakes classified as brown water lakes (15 to 50 TCU) and 28 lakes were considered highly coloured ($\leq 50~TCU$). Lake (excess) sulphate levels were also low (5.4 to 44.4 $\mu eq~L^{-1}$, x = 21.0) while organic anions ranged from 2.5 to 144.0 $\mu eq~L^{-1}$ (x = 41.4). Lake acidity reflected the inputs of both anthropogenic and natural acids.

The dilute and acid-sensitive nature of the south coast lakes was confirmed by various sensitivity indices including alkalinity, calcium, conductivity, the calcite saturation index (CSI) and bicarbonate to excess sulphate ratio. A deficit in alkalinity was apparent in all but two lakes, falling mostly in the 20 to 50 μ eq L⁻¹ range. A plot of south coast data on Henriksen's empirical acidification models suggests most lakes showing acidification effects are strongly influenced by natural organic acidity. The acid-base balance in lakes in the southwest corner of the island was more clearly related to anthropogenic sulphate levels. Both models are considered poor in their application to south coasts lakes due to the preponderance of weak (organic) acid dominated systems. The ratios of alkalinity to excess calcium and magnesium, excess sulphate to excess calcium and magnesium, and alkalinity to excess sulphate for the study lakes provides further evidence of extreme sensitivity to acidification. These ratios also provide evidence of ongoing acidification (alkalinity depletion). The Wright-Henriksen acidification equation was used to predict pH/alkalinity values for the study lakes, given specified scenarios of increases in sulphate deposition. The Wright-Henriksen model suggested a 50% increase in sulphate deposition could result in 32% of south coast lakes becoming acidified (pH <5.0, no alkalinity) while a doubling of current sulphate deposition rates could result in 64% of south coast lakes becoming acidified.

RÉSUMÉ

Scruton, D. A., and D. H. Taylor. 1989. A survey of dilute lakes on the south coast of insular Newfoundland in relation to natural and anthropogenic acidification. Can. Tech. Rep. Fish. Aquat. Sci. 1711: v + 58 p.

En juillet 1983 on a fait un relevé synoptique portant sur la qualité chimique de l'eau de 90 lacs de la côte sud de l'île de Tere-Neuve afin d'étudier leur sensibilité aux polluants atmosphériques venant de sources éloignées (TADPA) ainsi que les effets de ces polluants. Les lacs étudiés se caractérisent par leur très faible alcalinité (dans tous les cas, elle est inférieure ou égale à 200 μ éq·L⁻¹, x=23,7) et un pH se situant dans la zone d'acidité (dans la plupart des cas, il est inférieur à 6,50, x=5,76). La couleur des eaux va de claire à très opaque: 51 lacs se classent dans la catégorie des eaux brunes (de 15 à 50 UCR) et 28 dans la catégorie des eaux très opaques (plus de 50 UCR). La concentration de sulfate (en excès) est également basse (de 5,4 à 44,4 μ éq·L⁻¹, x=21,0), tandis que la concentration d'ions organiques va de 2,5 à 144,0 μ éq·L⁻¹ (x=41,4). L'acidité des eaux des lacs est le résultat des apports acides tant anthropiques que naturels.

Le caractère de dilution et de sensibilité à l'acide des lacs de la côte sud se confirme lorsqu'on examine divers indices de sensibilité intéressant notamment l'alcalinité, la teneur en calcium, la conductivité ainsi que l'indice de saturation en calcite (ISC) et le rapport bicarbonate-sulfate en excès. On constate que l'alcalinité est insuffisante dans tous les lacs sauf deux et qu'elle se situe généralement entre 20 et 50 μéq·L⁻¹. Si l'on applique les données recueillies au modèle empirique d'acidification de Henriksen, il ressort que la plupart des lacs où s'observent des signes d'acidification subissent fortement l'influence de l'acidité organique naturelle. On constate que dans les lacs de l'extrémité sud-ouest de l'île, l'équilibre acide-base est plus nettement lié aux apports de sulfate anthropiques. Ces deux modèles s'appliquent mal aux lacs de la côte sud car les acides faibles (organiques) prédominent. On constate une fois de plus l'extrême sensibilité des lacs à l'acidification lorsqu'on regarde les rapports suivants : alcalinité-calcium et magnésium en excès; sulfate en excès - calcium et magnésium en excès; alcalinité-sulfate en excès. Ces rapports témoignent aussi de la progression de l'acidification (baisse de l'alcalinité). On s'est servi de l'équation de l'acidification de Wright-Henriksen pour prévoir le pH et l'alcalinité des lacs étudiés dans l'hypothèse d'augmentations données des apports de sulfate : d'après les résultats, une augmentation de 50 % des apports de sulfate pourrait se traduire par l'acidification (pH inf. à 5,0, alcalinité nulle) de 32 % des lacs de la côte sud, tandis que si les apports de sulfate actuels doublaient, 46 % des lacs de la côte sud s'acidifieraient.

INTRODUCTION

The acidification of freshwater lakes and concurrent perturbation of resident biota due to the deposition of acidic precipitation is a well documented global phenomenon. Acidification damage has been reported from Scandinavia (Almer et al. 1974, 1978; Drablos and Tollan 1980; Overrein et al. 1980), the United Kingdom (Wells et al. 1986), Finland (Anonymous 1982), West Germany (Kraus et al. 1986) and other regions in (e.g. Freiesleben and Rasmussen 1986; et al. 1986; Wieting 1986). acidification in North America was first reported from the LaCloche Mountains in Ontario (Beamish and Harvey 1972) and the Adirondack Mountains in the northeastern U.S.A. (Schofield 1976). Acidification of freshwaters in northeastern North America has since become recognized as a more widespread problem (Harvey et al. 1981; Bangay and Riordan 1983; Environmental Protection Association 1983; Kelso et al. 1986; Jefferies et al. 1986; Linthurst et al. 1986).

Recent lake surveys in insular Newfoundland have confirmed the extreme susceptibility of the island's freshwaters to anthropogenic acidification but evidence of widespread acidification damage is lacking (Clair et al. 1982; Scruton 1983; Howell 1986; Scruton 1986; Howell and Brooksbank 1987). Sensitive freshwaters are demonstrating alkalinity depletion and possible slight pH reduction (Scruton et al. 1987) but the most acidic systems (pH \leq .0) appear to be weak organic acid dominated. The preponderance of organic acid dominated systems has made it difficult to determine the contribution of atmospheric deposition of strong acids (NO₃ - and SO₄ - 2) to the acid-base chemistry of the region's lakes. The absence of historical water chemistry for sensitive, low order lakes precludes any comparison through time as evidence of progressing acidification.

temporal and spatial variability in deposition of strong acids in precipitation is not well understood in insular Newfoundland. While the deposition scenario on the island is still unclear, sulphate values in lake water have suggested a west to east, or southwest to northeast, gradient in the atmospheric deposition of sulphate (Scruton 1983, Howell and Brooksbank 1987). Precipitation acidity in the Atlantic Region has varied between pH 4.0 and 6.0, with most values between 4.2 and 4.8 (Environmental Protection Service 1984), while rainfall pH (annual) in insular Newfoundland is considered to range between 4.5 and 4.9 (Anonymous 1983). date, severe acidification damage has not been demonostrated in global areas receiving a rainfall pH of 4.7 or greater (Schindler 1988). Sulphate deposition on the island averages between 15 and 20 kg of wet sulphate per hectare per year, and the south coast deposition levels are considered to approximate 20 kg ha 'yr 'so ' or greater. The Environmental Protection Service (1984) reported annual wet deposition of sulphate at Gander in Central Newfoundland to vary from a low of 19.2 (1977) to a high of 35.8 (1978) kg ha 'yr' over the period 1977 to 1980. From 1983 to 1986, wet sulphate deposition at Baie D'Espoir was between 10 to 15 kg ha-1 yr-1 (Phillips 1987). Twenty kilograms of wet sulphate per hectare per year is the target loading level the Canadian government has established to safeguard moderately sensitive ecosystems (Bangay and Riordan 1983). Extremely sensitive freshwaters, as in insular Newfoundland, may be susceptible to lower loading levels. Recent evidence suggests that this target level may be insufficient to protect extremely sensitive freshwaters (Schindler 1988), particularly those receiving a component of natural (organic) acid loading (Jones et al. 1986).

The south coast of Insular Newfoundland receives the highest annual precipitation in the province, ranging from 1500 mm in the east to over 2000 mm in the west (Banfield 1983). This amount of precipitation contributes to the expected higher rate of acid loading the region receives relative to other parts of the island. The high precipitation input also results in rapid runoff and high lake flushing rates, which are hydraulic characteristics that contribute to lake sensitivity and alkalinity depletion. Additionally, high precipitation coupled to low rates of evapotranspiration have resulted in a net moisture surplus in many regions of insular Newfoundland, contributing to humification of soils (Damman 1983).

In relation to the future emissions/deposition scenario in Atlantic Canada, concern has been expressed recently as to sulphur emissions arising from Nova Scotia Power's coal-burning electrical generation facility at Lingan on Cape Breton Island. This facility, while using state-of-the-art technology to control nitrogen emissions, does not significantly reduce its sulphur emissions, and utilizes local coal with a high sulphur content (1-8% by weight). Control technologies (combustion and Control technologies (combustion and post-combustion options) are unlikely to be implemented until the mid to late 1990's if at all (EPS 1984). The south coast is in close geographic proximity to (150 to 450 km), and downwind of, this utility, which represents the single regional point source of sulphur pollution of concern to the Island of Newfoundland. On a regional basis, emissions from coal fired generation in the Atlantic Region could grow from 120,000 MT to 200,000 MT over the next 10 to 15 years if no pre-combustion or emissions controls are implemented (Environmental Protection Service 1984). In addition to the predicted overall growth in coal-fired emissions in the Maritimes, the geographic focus of these emissions are to change. The Nova Scotia Power Commission plans to decommission several older coal burning power plants, with power production being replaced by increased coal consumption at newer plants, including Lingan. These regional considerations, in combination with geological sensitivity and high levels of precipitation, suggest the south coast of insular Newfoundland has potential for localized or widespread damage to freshwaters from the long range transport phenomenon.

Future trends in acid-causing emissions, and hence deposition rates, are difficult to forecast. There was a very clear downward trend in SO₂ emissions from the period 1970-73 to the early 1980's (1980-83) (Martin and Brydges 1986). The downward trend in U.S. emissions appears to have levelled out and may be slightly increasing, with projected slight increases over the 1980-85 to 1991 period. Eastern Canadian sulphur emissions

are forecast to increase slightly over the 1980-83 period to 1990, and then decline as emission control programs are implemented to achieve 1994 target levels. The North American deposition scenario, particularly the contribution of American emissions to acid deposition in Atlantic Canada, is particularly difficult to forecast.

For the purposes of this study the south coast of insular Newfoundland has been delineated as a large geographical area from 59°10'W latitude to 54°10'W latitude, encompassing major and minor river drainages from the Isle aux Morts River (west) to Piper's Hole River (east) but excluding the Burn Peninsula. This encompasses two major, ecologically unique regions identified for the island; the Maritime Barrens Ecoregion and the (southern) Long Range Barrens Ecoregion (Northlands Associates 1982). The Maritime Barrens Ecoregion is characterized by extensive barren lands with dwarf shrub heath vegetation, shallow acid soil cover, and poor to no forest development, except in some of the major river valleys. The underlying geology is mostly gneisses, and mafic volcanics; all low in available carbonates, and granites. consequently affording little buffing capacity to surface waters. It is thought the region originally contained some forest development which has been eliminated by fires and changing soil characteristics (31% of the land area was burned over between 1958 and 1974) (Damman 1983). fires and loss of forest cover have resulted in thick raw humus horizons in the soil with the organic matter contributing a natural colour and acidity to the freshwaters. This ecoregion is characterized climatically by cold summers, mild winters, a high frequency of fog, and discontinuous winter snow cover due to frequent winter rainfalls.

The Long Range Barrens Ecoregion, in the southwest corner of the study region, is characterized by highlands containing extensive "tuckamore" (coniferous shrub thickets). This ecoregion contains fewer bogs (mostly of slope and basin type) than the Maritime Barrens Ecoregion. The region is further characterized by high elevations (300-600 m) and extremely high precipitation inputs (1800-2000 mm per yr). Climatically, the region is cold with permanent snow cover through the winter months.

The Department of Fisheries and Oceans, recognizing acidification as a significant threat to fish and fish habitat in Canada, initiated a departmental program on aquatic effects associated with acid precipitation in 1981. A National Inventory Survey (N.I.S.) of lakes in acid-sensitive terrain was undertaken initially to evaluate the inter- and intra- regional differences in sensitivity and responses to anthropogenic deposition of strong acids (Kelso et al. 1986). The inventory program has been subsequently conducted in all Eastern Canadian provinces, excepting P.E.I. In 1981, 109 lakes across the island of Newfoundland were surveyed for morphometry, chemistry, and biota, while a survey of 130 lakes in Labrador was completed in 1982 as part of the NIS (Scruton 1983, 1984).

The original island-wide survey of insular Newfoundland lakes in 1981 pointed to the south coast of the island, particularly the southwest

corner, as having the considered potential for damage of freshwaters due to the long range transport of air pollutants. The N.I.S. survey (Scruton 1983) and subsequent regional analysis of larger data bases (Jefferies et al. 1986; Howell and Brooksbank 1987; Schindler 1988) also identified the highlands in central Newfoundland and the eastern side of the northern Peninsula as having highly susceptible freshwaters. Lakes on the south coast, surveyed in N.I.S., demonstrated low alkalinities (all $\leq 100~\mu eq~L^{-1}$), low pH values (71% less than pH 6.0), and the considerable influence of natural organic acids. However, due to the broad-scale nature of the 1981 survey, only 24 lakes were surveyed in the south coast region. Fisheries and Oceans, in consultation with the Newfoundland Acid Rain Technical Group (NARTG) (a group of researchers active in aquatic, terrestrial, and atmospheric monitoring of acid rain effects), decided a second synoptic survey was warranted, geographically scoped to the south coast, to more fully address the sensitivity and current status of lakes in this region in relation to acidic deposition. Further, this survey was designed to sample lakes in approximation to their natural distributions with respect to key morphometric characteristics (area, drainage order), to allow for representative extrapolation to all of the region's lakes.

METHODS

Lakes selected for survey were located along the south coast of insular Newfoundland from Port aux Basques (west) to Swift Current (east), and were within major and minor river watersheds draining to the south. All lakes were located in remots, undisturbed areas removed from population centers and point sources of pollution, in geologically acid-sensitive terrain. Lake selection was biased to sample more lakes in the southwest coast of the island (to the west of 58°W latitude), an area acknowledged to receive the highest annual precipitation in the province (Banfield 1983) and consequently suspected of receiving the highest rate of acid loading in the region.

Lake selection was stratified in relation to lake size and drainage order. Lakes in the 0 to 10 ha size class were not sampled in the previous island-wide lake survey (Scruton 1983) due to limitations in access by fixed-wing aircraft. This lake size was subsequently identified as the most abundant size class in insular Newfoundland (Dobrocky Seatech Nfld. Ltd. 1983), and consequently survey efforts were directed to include a number of lakes of this size class in this study. Similarly, the natural distribution of lakes by drainage order was known and lake selection was stratified to approximate natural distributions.

A total of 116 lakes were sampled between July 18 and 24, 1983. Ninety (90) of these lakes were sampled for the first time (600 series) while 26 lakes (200 series) were resampled from previous inventory and/or monitoring networks. Data from the resampled lakes contribute to a larger, active data set and will not be addressed (in detail) in this report.

Water sampling was conducted at the visually estimated lake midpoint from the floats of a helicopter. Water samples were collected from each lake by dipping sample bottles 0.5 m below the lake surface. A 250-ml bottle was filled for analyses of selected parameters at a field laboratory while a 1-liter bottle was filled for detailed laboratory analyses. A pre-acidified (0.25 ml concentrated nitric acid) 125-ml bottle was filled for aluminum analysis.

Samples were analyzed at a field laboratory for pH, alkalinity, and conductivity within 24 h of collection. Alkalinity was determined by Gran titration (after the Ontario Ministry of Natural Resources 1980) and subsequent calculation from the data set (Kramer 1978). Field pH values were the initial pH reading of the Gran titration. Conductivity was measured using a Fisher Model 152 conductivity meter, at ambient temperature, and later adjusted to 25°C.

Samples were shipped to a selected analytical laboratory (Oceanchem Ltd., Halifax, N.S.) upon completion of the sampling program. Water samples were analyzed for 17 separate parameters while an 8 additional parameters were calculated. A list of analytical procedures, equipment, and limits of detection is provided in Table 1. All methods followed those outlined in Environment Canada (1979) or American Public Health Association et al. (1975).

Analytical methods employed in this study compared favorably with those utilized in the previous N.I.S. surveys (Scruton 1983, 1984) with the following exceptions. Major anions (including chloride, sulphate, and nitrate) were determined by ion chromatography in this study while previously, these parameters had been determined colourimetrically by auto analyzer. Sulphate was also determined by the methyl-thymol blue (MTB) method to allow comparisons between values for both methods. The MTB method, with colourimetric determination, is recognized to overestimate sulphate in highly coloured waters (Kerekes and Howell 1983). Ion chromatography is now the preferred method for sulphate analysis in moderate to highly coloured waters.

Total hardness was calculated by summing determinations for calcium and magnesium. Bicarbonate was calculated from field alkalinity values and pH. Non-marine or "excess" concentrations of calcium, magnesium, sodium, potassium, and sulphate were calculated on the basis of the ratio of these ions to chloride in saltwater, assuming all chloride in each sample was of marine origin. The measured level of chloride in each sample is used to determine the marine contribution of each ion which is then subtracted from the measured values (Thompson 1982). Organic anions (COOH) were calculated from dissolved organic carbon (DOC) and pH after the method of Oliver et al. (1983).

Morphometric characteristics of the study lakes (lake surface area, watershed area, ratio of watershed to lake area, elevation, and distance from the sea coast) were determined from 1:50,000 or 1:250,000 scale maps. Other morphometric characteristics such as bathymetry, estimated maximum/mean depth, secchi depth, etc., usually

evaluated in the field, were not determined in this study.

Calcite saturation index (CSI) values, as a sensitivity indicator, were calculated after the following equation (Thompson and Hutton, 1982):

 $CSI=log(Ca^{+\frac{1}{2}})+log(HCO_{1}^{-})+pH-logK_{2}-logK_{SP}$

where Ca⁺², HCO, are molar concentrations, log K,

(disassociation constant for carbonic acid) = -10.3, K_{SP} (disassociation constant for calcite) = -8.3, and no corrections for temperature or ionic strength were made.

RESULTS

GENERAL LAKE CHARACTERISTICS

For the purposes of sorting and describing the study lakes, three sub-regions were delineated within the confines of the south coast. Forty-three (48%) of the study lakes were located in the Southwestern sub-region (west of 58°25'W longitude), 35 lakes (39%) in the Central sub-region (between 58°25'W longitude and 56°15'W longitude) with the remaining 12 lakes (13%) being located in the Eastern sub-region (east of 56°15'W longitude).

Lakes were sampled from four major, broadly distributed, geological types underlying the south coast region (Rogerson 1983) including granites (43 lakes, 48%); gneiss, schist and foliated granites (26 lakes, 29%); gabbro, diorites, etc. (5 lakes, 6%); and acid to mafic volcanics, siltstones, sandstones, metamorphics, etc. (16 lakes, 18%). Sixty-nine lakes (77%) located on relatively insoluble bedrock deemed highly sensitive to acidification (granites, greisses), while the remaining 21 lakes were located on bedrock classified as of moderate sensitivity (Schilts 1981). The recently updated geological of Insular Newfoundland map (Newfoundland and Labrador Department of Mines and Energy, geological map of insular Newfoundland, 1986) has revealed considerable geological heterogeneity within the study area. Despite the relatively small watershed size for most lakes, 27 lake watersheds (30%) were underlain by two different geotypes, while 6 lake catchments (7%) were underlain by three major geological formations. Twenty-four lakes were underlain by combinations of granites and gneiss/schist, all acid-sensitive, while 9 lakes underlain by combinations of geotypes with different (moderate to high) sensitivities. For these nine lakes, geological sensitivity WAS assigned, by convention, on the basis of the geotype contributing of the largest area in the watershed. Surface waters which drain bedrock with different capacities to reduce acidity will generally reflect the character of the most dissoluble bedrock (Potter et al. 1982). In this study all geotypes are moderately/highly sensitive and carbonate poor. No attempt was made to assign sensitivity on the basis of the most dissoluble bedrock type, for the watersheds with complex bedrock geology.

Lake surface area varied from 3.0 to 773.5 ha $(\bar{x}=62.0)$ with 26 lakes (29%) less than or equal

to 10 ha, 51 lakes (57%) from 10.1 to 100 ha, and 13 lakes (14%) from 100.1 to 1000 ha in size (Fig. 2, Table 2). Watershed area varied from 15.4 to 5160.0 ha $(\overline{x}=421.0)$ with most lake watersheds less than 1000 ha (82 or 91% of the lakes) (Table 2). The watershed area to lake area ratio (WA:LA), a convenient index for evaluating hydrologic control (flushing rates) of lake susceptibility to acidification, ranged from 1.0 to 87.7 $(\overline{x} = 13.7)$ (Fig. 2, Table 2). Fifty-six lakes (62%) had low ratios of less than 10:1, while only 8 lakes (9%) had ratios in excess of 25:1. Lakes sampled in this survey ranged from true headwater (first order) lakes to larger lakes lower in the drainage system (fifth or higher order), and also included three ponds with no apparent outlet (possibly seepage lakes). Lake distribution by drainage order is as follows: first order - 50 lakes (56%), second order -20 lakes (22%), third order - 7 lakes (8%), fourth order - 5 lakes (6%), and fifth (or higher) order - 8 lakes (9%) (Fig. 2, Table 2). Lake elevation varied from 30 to 495 m above sea level (x = 301.0). Only one lake had an elevation of less than 100 m (Lake #600, 30 m), while 50 lakes (56%) were located between 100.1 and 300 m, with the remaining 39 lakes (43%) found at higher elevations (300.1-500 m) (Fig. 2, Table 2). The of the study lakes from the coast distance (nearest salt water), a variable which has proven valuable in assessing marine influence on chemistry, ranged from 2.5 to 80.0 km (\bar{x} = 29.1). Fifty-six (62%) of the lakes were within 30 km of salt water (Table 2).

Water colour, which has previously been demonstrated to be a good correlate of organic content (Scruton 1983, 1984), was found to range from 3.5 to 120.0 T.C.U. ($\overline{\chi}=47.7$) (Fig. 3, Table 2). Using criteria previously established to classify lakes as to colour, 10 lakes (11%) are considered clearwater systems (0 to 15 T.C.U.), 51 lakes (57%) were deemed brown water systems (16 to 50 T.C.U.), while 28 lakes (31.5%) were classifed as highly coloured (\geq 50 T.C.U.) (Fig. 2). Turbidity values in the study lakes ranged from 0.06 to 4.50 JTU ($\overline{\chi}=0.77$). The majority of the values were in the 0.50 to 1.00 J.T.U. range, while 13 lakes (15%) had turbidities in excess of 1.0 (Table 2).

Нq

pH values for the study lakes, measured in the field, varied from 4.61 to 6.67 ($\vec{x} = 5.76$) (Table 2). Six lakes (7%) had pH values of less than 5.00, 23 lakes (27%) had values between 5.01 and 5.50, 23 (27%) lakes with pH values from 5.50 to 6.00, 22 lakes (26%) with values from 6.01 to 6.50, while 11 lakes (13%) had values in excess of 6.50 (Fig. 3). The pH distribution was unimodal with relatively even distribution across pH classes 5.0 to 5.5, 5.5 to 6.0, and 6.0 to 6.5. Clearwater lakes had a pH range of 5.31 to 6.44 and the 23 lakes below pH 5.31 were either brown water (8 lakes) or highly coloured (15 lakes). The spatial distribution of pH values is presented in Fig. 5.

ALKALINITY

Alkalinity values, as determined by potentiometric titration and Gran analysis, varied

from -29.6 to $154.8~\mu eq~L^{-1}~(\vec{x}~\approx~23.7~\mu eq~L^{-1})$ (Table 2). All study lakes had alkalinities well below 200 $\mu eq~L^{-1}$ (10.0 mg L^{-1}), adopted by most researchers as standard for differentiation of high to moderately sensitive freshwaters from relatively non-sensitive systems (Ontario Ministry of the Environment 1981, Harvey et al. 1981, Bangay and Riordan 1983). Fourteen lakes demonstrated zero or negative alkalinities and, by definition, were considered acidified. Sixty-six lakes (73%) had alkalinities of less than 40 $\mu eq~L^{-1}$ and are considered extremely (critically) sensitive to potential acidification (Fig. 3). The spatial distribution of alkalinity values is presented in Fig. 5.

Several authors have decided to adopt the stringent definition of acidification as complete loss of alkalinity (e.g. Harvey et al. 1981, Ontario Ministry of the Environment 1981). Recently, other researchers have considered lakes which have lost alkalinity, but still retain some buffering capacity, as acidified (e.g. Schindler 1988). In this report we consider lakes with appreciable alkalinity losses, but with some residual buffering capacity, as becoming acidified while lakes with exhuasted alkalinity are considered acidified.

CONDUCTIVITY

Specific conductance, or conductivity, for the study lakes ranged from 7.2 to $76.0~\mu \rm Scm^{-1}$ ($\overline{x}=14.0$) with 78 lakes (88%) having extremely low conductivities of less than $20~\mu \rm Scm^{-1}$ (Table 2). Seven lakes (8%) had conductivities from 20 to $30~\mu \rm Scm^{-1}$ while only four lakes (4%) exceeded this value. Conductivity was positively correlated (Pearson product-moment correlation here and elsewhere) to the sum of anions (r = 0.36, sum of cations (r = 0.25), chloride (r = 0.26) and bicarbonate (r = 0.23), as well as the base cations, calcium (r = 0.18) and magnesium (r = 0.21).

CATIONS

The ionic composition of the lakes can best be evaluated in relation to the order of ionic dominance (on an equivalent basis) (Table 3). Sodium was the dominant cation in 80 lakes (90%) with calcium dominant in 9 lakes (10%). In sodium dominated lakes, the catonic order was typically Na > Ca > Mg > K (70 lakes, 79%) or Na > Mg > Ca > K (10 lakes, 11%). Sodium levels varied from 30.7 to 113.1 μ eq $\frac{1}{1}$ (\overline{x} = 65.1), calcium from 6.8 to 158.8 μ eq $\frac{1}{1}$ (\overline{x} = 40.5), magnesium from 10.3 to 60.3 μ eq $\frac{1}{1}$ (\overline{x} = 23.3), while potassium values were low, ranging from 0.7 to 7.9 μ eq $\frac{1}{1}$ (\overline{x} = 3.7).

Considering non-marine . ('excess') concentrations of the major cations, calcium was dominant in 62 lakes (70%), sodium dominant in 22 lakes (24%) and magnesium dominant in 5 lakes (6%). Potassium ranked lowest in equivalence of these four non-marine cations in all lakes excepting one (#600). Non-marine calcium dominated lakes were underlain by both highly sensitive granites/gneisses (48 lakes, 54%) and volcanics/sandstones (19 lakes, 21%). Non-marine sodium dominated lakes were located exclusively on

granitic/gneitic bedrock while magnesium dominated lakes were underlain by the volcanics/sandstones geotype only. The range in non-marine concentrations of the major cations was as follows: non-marine calcium (5.7 to 157.1 μ eq L⁻¹, $\overline{x}=38.9$), non-marine sodium (0.0 to 47.1 μ eq L⁻¹, x=23.2), non-marine magnesium (6.9 to 56.5 μ eq L⁻¹, $\overline{x}=18.3$) and non-marine potassium (0.0 to 6.5 μ eq L⁻¹, $\overline{x}=2.2$).

Hydrogen ion concentration (H^+) for the study lakes varied from 0.21 to 24.5 μ eq L^{-1} (x = 3.6). Hydrogen ion did not contribute significantly (10% or greater) to the total cation sum except in lakes with pH less than 5.00. Hydrogen ion was dominant to potassium in 31 acidic lakes (34%) and was the second most important cation (after sodium), being dominant to the base cations (Ca+ 2 and Mg+ 2), in two extremely acidic lakes (#672, pH of 4.82; #687, pH of 4.61).

ANIONS

Chloride was the dominant anion in 68 lakes (84%), while organic anions (COOH) were dominant in 8 lakes (10%) and bicarbonate dominant in 5 (6%) (Table 2). Considering non-marine lakes concentrations only, organics were the dominant anion in 45 lakes (56%), bicarbonate in 22 lakes and excess sulphate in 14 lakes (17%). Organic anion dominated lakes were found in all geotypes excepting the gabbros/diorites/silstone/ sandstone group, while non-marine sulphate dominated lakes were found mostly in the highly sensitive granitic and gneitic bedrock. Four of the five gabbro/diorite/silstone/sandstone lakes were bicarbonate dominated. The abundance of organic acid-dominated lakes stresses the importance of quantitative determinaton of this ionic constituent in order to evaluate its role in the acid-base chemistry of dilute, bicarbonate poor systems.

Organic acid concentration (COOH) was calculated from dissolved organic carbon (DOC) and pH, after the method of Oliver et al. (1983). Dissolved organic carbon (DOC) varied from 0.3 to 20.0 mg L $^{-1}$ (x = 4.9) while organic anion equivalence (COOH) was determined to range from 2.5 to 144.0 (x = 41.4). Organic anion concentrations were less than 50 μ eq L $^{-1}$ in 67 lakes (83%), between 50 and 100 μ eq L $^{-1}$ in 9 lakes (11%) and exceeded 100 μ eq L $^{-1}$ in only 5 lakes (6%). Dissolved organic carbon was found to be significantly correlated with water colour (r = 0.26, p = 0.029, n = 85), although the regression coefficient is not as good as it has been in previous data sets (Scruton 1983, 1984).

Chloride values ranged from 39.5 to $166.4~\mu {\rm eq}~{\rm L}^{-1}~(\overline{\rm x}=75.4)$. All study lakes were located remote from human habitation and distant from the two major highways (Burgeo and Baie D'Espoir highways) transecting the study area. Chloride inputs from road salts and/or dust control products is not suspected. Additionally, there is no evidence of salt-bearing bedrock on the south coast (Newfoundland and Labrador Mines and Energy, Geological Map of Insular Newfoundland, 1986), consequently all chloride is assumed to be of marine origin, deposited in precipitation, as aerosols or in fog. On the basis of this assumption, the proportional

contribution of major ions (Ca⁺², Mg⁺² Na⁺, K+, SO₄⁻²) of marine origin can be calculated from the ratio of these ions to chloride in salt water and measured chloride values in each lake (Thompson 1982). The relative influence of the marine environment on the major ion chemistry of south coast lakes, expressed as proportional (average) marine contribution, is as follows: chloride (100%), sodium (64%), potassium (41%), sulphate (33%), magnesium (21%), and calcium (4%) (see also Table 5).

Sulphate, as determined by ion chromatography (IC) varied from 12.5 to 62.5 μ eq L $^{-1}$ (\overline{x} = 31.5), while non-marine concentrations ranged from 5.4 to 44.4 μ eq L $^{-1}$ (\overline{x} = 21.0) (Fig. 3, Table 2). Most of the lakes (79 lakes or 89%) had non-marine sulphate values in the 10 to 40 μ eq L $^{-1}$ range. Sulphate was also determined colourimetrically (by MTB), for comparison to previous surveys and also to quantitatively evaluate the interference of water colour in measurement. Values for MTB sulphate ranged from 16.7 to 60.0 (\overline{x} = 39.6). Values determined by both MTB and IC methods were highly correlated (r = 0.84, P < 0.001, n = 84) with MTB values exceeding IC values 82% of the time. The residual (MTB.SO $_{4}^{-2}$ - IC.SO $_{4}^{-2}$) was correlated (r = 0.24, P < 0.05, n = 84) to water colour providing evidence of colourimetric interference.

Bicarbonate values for the study lakes, as calculated from alkalinity, varied from 0.0 to 188.5 μ eq L⁻¹ ($\bar{x}=31.3$) with the 14 lakes with 0 or negative alkalinities having, by convention, no available bicarbonate.

ALUMINUM

Total aluminum in the study lakes varied from 33.0 to 554.0 μg L $^{-1}$ ($\overline{x}=357.0$) with most of the values (50 lakes, 56%) falling in the range of 350 to 450 μg L $^{-1}$ (Table 2). Only four lakes had values exceeding 450 μg L $^{-1}$. Aluminum was found to be correlated (negatively) to the base cations calcium (r = -0.33, p < 0.05, n = 85) and magnesium (r = -0.24, p < 0.05, n = 85) but was not well correlated to either lake pH (r = -0.17, p = 0.12, n = 80) or water colour (r = 0.12, p = 0.26, n = 85).

DISCUSSION

It is initially important to highlight that this survey was conducted during a season historically asociated with seasonal lows in the hydrological cycle of many of insular Newfoundland's freshwaters (Environment Canada 1985). The hydrological records (Environment Canada 1984) for Isle aux Morts River, Grandy Brook and Baie du Nord River, three gauged rivers within the study area, were reviewed with respect to water flow conditions prior to, and during, the survey. The records indicate that the early summer months of 1983 were extremely dry, with the most of June being the driest for the year for all three systems. A small spate preceded the survey period with flows subsiding as the survey progressed. Average daily flows during the study period were 20-40% less than the mean daily flows for the year (1983).

Low points in the hydrological cycle are coupled to seasonal highs in base cations and bicarbonate concentrations (and consequently pH), due to a greater contribution of groundwater (base flow) to freshwater chemistry (Skartveit 1981). flow is usually higher in dissolved constituents than runoff due to increased contact time between soils/rock and groundwater (Hem 1970). Similarly, sulphate and organic content (and its correlates) experience seasonal lows due to vegetational uptake and retention in the terrestrial confines of the watersheds (Dovland and Semb 1978). Consequently pH, alkalinity, and base cation data from this survey would be considered to represent a "best case" scenario with respect to an evaluation of acid sensitivity and acidification effects. Conversely, sulphate and organic content (colour, DOC, COOH) data may not adequately characterize, in quantitative terms, anthropogenic and natural sources of acidity, due to considerations of seasonality and hydrology.

Ryan and Wakeham (1984) have shown seasonal variation in Newfoundland lakes may not necessarily follow the hydrological cycle, although their sampling regime was insufficient to make direct comparisons. Longer term and more frequency data records on some Newfoundland rivers (unpublished NAQUADAT data; Scruton 1984) do support the aforementioned assumptions, given an examination of the hydrological records for guaged rivers within the study area in 1983.

LAKE AREA AND DRAINAGE ORDER

For the purposes of evaluating the representativeness of the lakes selected for survey, the distribution of the study lakes, by area and drainage order, is compared to natural distributions (Fig. 4) as determined by extensive counts and measures (digitization of all lakes > 1 hectare) of lakes and rivers along the south coast of Newfoundland (Dobrocky Seatech Nfld. Ltd. 1983). It is apparent that the lake size classes 0-10 ha and 100-1000 ha were underrepresented in this survey while the 10-100~ha size class was overrepresented. The 0-10~ha size class was not sampled in proportion to natural distributions for variety of reasons. An underlying objective of this survey was to evaluate lake acidification potential in relation to productive fish habitat. The majority of these smaller ponds can be characterized as extremely shallow (3 m or less), bottomed with abundant macrophytic soft vegetation, often with intermittent or no outlet, and frequently containing no potential reproductive substrates in the littoral zone or in tributaries. These lakes are considered to have limited potential for fish production regardless of their chemical status. Conversely, lakes in the 10 to 100 ha size class were oversampled, again because of their fish production potential. Smaller lakes frequently contain a higher proportion of littoral zone relative to larger lakes, and therefore would have a greater fish production potential per unit area as per considerations expressed by the morphoedapic index (Ryder 1965). Not withstanding the above, small ponds do contribute to fish production in insular -Newfoundland, and are valuable nursery areas for juvenile Atlantic salmon (Pepper 1976).

Similarly, the comparison of the study lake drainage orders and natural distributions indicates that the study lake sample is reasonably representative of natural drainage orders (Fig. 4). The previous island-wide (1981) survey had been highly biased to first order, headwater lakes (see also Fig. 4), largely because of the acknowledged high sensitivity of this lake type to acidification (Harvey et al. 1981). In this survey no biases were imposed (in relation to lake order) so as to evaluate lake drainage order as a contributing factor to sensitivity and the acidification process.

CATIONS

The relationship between the two principal cationic products of unstressed geochemical weathering, calcium and magnesium, is important in evaluating the geological controls on chemistry and buffering capacity against a scenario of atmospheric acid loading. Concentrations of these two cations were extremely low relative to other regions of Canada (Kelso et al. 1986; Jefferies et al. 1986; Howell and Brooksbank 1987). Within the island of Newfoundland, Howell and Brooksbank (1987) have reported excess calcium and magnesium concentrations to be lowest along the south coast and the eastern side of the Northern Peninsula. The non-marine concentration of these two cations were highly correlated (r=0.59, p 0.001, n=89), as were correlations of calcium (r=0.49, p 0.001, n=75) and magnesium (r=0.47, p 0.001, n=75) to bicarbonate, indicating similar lithological sources for these parameters. Non-marine calcium was dominant to non-marine magnesium in all but 5 lakes, and the Ca:Mg ratio varied from 0.18 to 5.72 ($\overline{x} = 2.20$) with 70 lakes (79%) falling in the range of 1.0 to 3.0. The magnesium dominated lakes (n = 5) were characteristically higher order (3°, 5°) with magnesium values well above the mean for the data set. Lakes with the higher Ca:Mg ratios (exceeding 3.0, 14 lakes) were characterized as predominantly headwaters (1° or 2° order, n = 12, 86%) with pH values in excess of 6.0 (n = 9, 64%). Comparatively, lakes in the island-wide survey underlain by acid-sensitive geotypes demonstrated a narrower range is Ca:Mg ratio, 1.2 to 2.7 (x = 2.0) (Scruton 1983). The non-sensitive, (x = 2.0) (Scruton 1983). The non-sensitive, hardwater, high pH lakes in the island-wide survey demonstrated much greater calcium dominance (Ca:Mg ratio of 25:1 for limestones, 17:1 for conglomerates) reflecting the availability of calcium carbonates.

The dominance of non-marine sodium in 22 of the lakes, and the apparent importance of 'excess' sodium as the second most important cation in ('excess') calcium dominated lakes (43 lakes) is consistent with other survey data, including the N.I.S. (Scruton 1983). Howell (1986) has previously reported 26 of 34 (76%) Environment Canada's (Inland Waters Directorate or IWD) LRTAP monitoring lakes to be sodium dominated with the remaining 8 lakes calcium dominated.

In this study, sodium dominated lakes were most commonly underlain by granites and other silicous bedrock types. The bedrock geology and topography of the south coast does not suggest widespread sodium bearing bedrock nor the likelihod of extensive distribution of marine

The stoichiometry between concentrations of sodium, calcium, and magnesium in this, and the 1981 survey, does suggest a geochemical source for the ion. The average mean non-marine sodium for this study (23.0 peg L similar to values for lakes underlain by granites (x = 32.3, n = 45), gneisses (x = 20.9, n = 17) and acid volcanics (x = 28.7, n = 17) in the previous island-wide survey (Scruton 1983). It is possible sodium was overestimated in the laboratory or there was insufficient sea salt correction for non-marine cations (i.e. chloride was underestimated). Howell (pers. comm.) believes that researchers in Atlantic Canada may not be making sufficient sea salt conection for aerosol contribution of sodium to freshwater chemistry. However, in surveys of this type, we must assume that marine contribution of sodium is in the same proportion as the NA:Cl ratio in salt water. Quality assurance checks, however, did not indicate any apparent biases in measurement of these two parameters.

possible There two additional explanations for the importance of non-marine sodium in the chemistry of south coast lakes. Thompson (1982) has provided evidence of cation exchange, primarily marine sodium for calcium, on a south coast river (Isle aux Morts River), suggesting this phenomenon may also be occurring in other dilute coastal rivers in Atlantic Canada. This type of cation exchange would upset the normal ionic sequence expected from purely geochemical weathering. Overrein et al. (1980) and Seip and Tollan (1982) have also investigated ion exchange reactions in Norwegian rivers involving sodium for hydrogen ion (H+), as a marine contributor to seasonal (episodic) acidification. Sullivan et al. (1988) also recently investigated this phenomenon in freshwaters along the Atlantic coast the U.S.A. and found no evidence, on a broadscale synoptic basis, of aerosol deposition in the acidification process.

In reviewing the characteristics of the non-marine sodium dominated lakes, 16 of 22 (73%) are within 25 km of the coast. These lakes are underlain by highly sensitive geotypes (granites, greisses) and consequently are characterized by very low alkalinities (-29.6 to 45.6 μ eq L⁻¹, $\bar{x}=4.0$) with 8 (36%) lakes having 0 or negative alkalinities. Sodium dominance in these lakes is largely a reflection of the extremely low base cation (calcium and magnesium) concentrations, rather than indicative of elevated sodium levels.

A major concern as to the importance of non-marine sodium as a major cation in many lakes is whether this is, in fact, evidence of significant geochemical supply. Assessments of sensitivity and acidification status (using empirical models) employ the underlying assumption that base cation concentrations (calcium and magnesium) provide a measure of "original" buffering capacity (HCO, concentrations) prior to titration by strong acids in precipitation. If there is a significant geological source for sodium, and sodium is weathered in proportion to bicarbonate, this assumption is not met. In this instance, original alkalinity would be underestimated as would the amount of acidification (as loss of alkalinity). Wright (1983) has suggested that sodium dominated lakes, be it a result of analytical error or geochemical

supply, do not meet the assumptions implicit in development of many empirical acidification models.

MARINE INFLUENCE

Previous studies have confirmed considerable influence of the marine environment on the chemistry of soft water lakes in insular Newfoundland (Scruton 1983). The proportional marine contribution of the four major cations and two major anions from recent and extensive survey programs is compared in Table 4. The proportional contribution of marine ions to south coast lake chemistry compares favorably with a river survey, conducted in 1981-82 (Scruton 1986), which included a large component of south coast rivers (and was also adjusted for temporal variability). In relation to the island-wide survey of 1981 (Scruton 1983), proportional contribution of sea salts to south coast chemistry is higher for Ca¹², Mg², Na¹, SO₄², and lesser for K¹. This could, in part, be explained by seasonality differences in the two sampling schedules but it is more likely a consequence of the proximity of south coast lakes to salt water (average distance from the coast is 29.1 ka). The preponderance of southerly and southwesterly air flows on the south coast and the topography of the region also contribute to higher proportional marine deposition. In addition, the island-wide survey included a number of highly mineralized (circumneutral and alkaline) lakes and the proportional influence of ions of marine origin on the chemistry of these lake types would be greatly

Ions from salt water can be deposited inland via aerosols, dry deposition, and in precipitation and fog. The magnitude of deposition is related to factors including distance of a water body from salt water, wind and precipitation patterns, evapotranspiration rates and topography. Sea salts fall out rapidly in dry deposition or are quickly scavenged by precipitation, frequently resulting in a sharp coastal gradient (Wright et al. 1977, Wright and Henriksen 1978). The two principal ions associated with salt water, chloride and sodium, demonstrated negative correlations to the distance from salt water $((r = -0.38 \text{ and } -0.37, P \le 0.001, respectively})$ confirming this expected coastal gradient. There was no evidence of topographic influence on deposition of marine salts, which is not surprising owing to the rather uniform elevation of the study lakes (excepting lakes in the Long Range Mountains) and precipitous nature of the south coast and major river valleys. Howell (1986), in evaluating the chemistry of 34 Environment Canada (IWD's) LRTAP monitoring lakes, found coastal Newfoundland lakes to show little marine aerosal effect, suggesting the high altitude of these monitoring lakes, minimizes the marine influence. This is in contrast to the NIS lakes surveyed in 1981 (Scruton 1983) and this data, where the proportional contribution of marine aerosols to freshwater chemistry is readily apparent and the sharp coastal gradient in the two principal ions deposited as marine aerosols, sodium and chloride, is well established.

рΗ

pH of freshwaters involves the The interaction of a number of acid-base reactions dissolution, precipitation, reduction-oxidation, adsorption-desorption, photosynthesis, decomposition of organic matter, etc. Precipitation (including chemical composition, acidity, and quantity) is just one factor influencing lake acidity. Lake pH was correlated with a number of morphometric characteristics including lake surface area (r = 0.22, P < 0.05), watershed area (r = 0.35, P < 0.001) and the WA:LA ratio ((r = 0.25, P < 0.001))P <0.001) and the WA:LA ratio ((r = 0.25, P <0.05), confirming the importance of catchment area in determining the availability of buffering agents and lake area in governing the hydrological interactions between buffering capacity and deposition rates (i.e. through flushing rates). Lake acidity (pH) was also very highly significantly correlated (P <0.001) with field alkalinity ((r = 0.77) and with the base cations, calcium (r = 0.47) and magnesium (r = 0.45), therefore highlighting the importance of geochemical weathering, moderated by watershed processes, in the acid-base control of the lakes.

insular Newfoundland, natural organic acidity is a major contributor to the acid-base balance of freshwaters owing to the dilute nature of most systems, and the preponderance of peatlands and organic soils across the island (Scruton 1983, 1985; Howell and Brooksbank 1987; Earle et al. 1987). Water colour was also correlated to pH (r = -0.34, P $\langle 0.001$), pointing to the role of organics in pH control across the range from clear to highly coloured study lakes. Clearwater lakes in this study had a pH range from 5.31 to 6.44 ($\overline{x} = 5.82$), while in the previous island-wide survey all clearwater lakes demonstrated a pH of greater than 5.5 (Scruton 1983). Organics are an important contributor of H ion to freshwater and, in extremely dilute poorly buffered systems, will significantly contribute to pH declines in proportion to their concentration. The importance of colour, as a correlate of organic content, in determining the pH of softwater lakes in insular Newfoundland and Labrador has been previously demonstrated (Scruton 1983, 1984). It is important to note that pH did not correlate with sulphate ((r = -0.11, P = 0.34, n=84) or non-marine sulphate (r = -0.10, P = 0.36, n = 84) as indicators of anthropogenic influences on lake acidity. Multivariate analysis of the N.I.S. data (Earle et al. 1986) also did not identify a 'LRTAP factor', associated with pH, as a major determinant of chemical variation in Newfoundland lakes.

Several authors have demonstrated a bimodal pH distribution in acid sensitive regions undergoing high acid loadings and have suggested this type of distribution is characteristic of a seriously impacted region (Almer et al. 1974; Schofield 1976; Wright and Gjessing 1976; Machell et al. 1983). This bimodal distribution is consistent with bicarbonate titration by a strong acid, yielding lakes with intact buffering regimes (pH 6.0 and greater) and lakes with serious depletion or loss of buffering capacity (pH 5.5 and less). Few lakes are located in the transition zone (pH 5.5 to 6.0) as this part of the titration curve is steep, subject to rapid pH decline in response to acid loading. The pH

distribution of south coast lakes is fairly even across the pH range 5.0 to 6.5. This would suggest that south coast lakes, assumed to be receiving the highest anthropogenic input of acids in the province, have not received sufficient loadings to have titrated most of the available buffering capacity. Lakes are equally represented in the pH range suggesting intact buffering systems (pH >6.0), in the steep portion of the pH-alkalinity titration curve (pH 5.5 to 6.0), as well as in the acidic range (pH \leftilde{5}.5).

The previous island-wide survey demonstrated a much wider pH range (4.90 to 8.39) reflecting a sample which included a number of moderately well buffered lakes in central and western Newfoundland, to hardwater alkaline lakes, underlain by carbonate bearing sedimentary rocks, on the western side of the Northern Peninsula. The modal pH class in the island-wide survey was 6.0 to 6.5. The considerably narrower pH range of south coast lakes is a primarily reflection of the surficial geology as all lakes are underlain by the highly sensitive igneous formations (granites, gneisses) and moderately sensitive volcanics and sandstones, all low in readily weatherable HCO₃ /Ca⁺²/Mg⁺². Natural and anthropogenic acid loading are also contributing to lower pH values in south coast lakes.

ALKALINITY

Adopting the Ontario Ministry of the Environment's (1981) classification system for lake sensitivity to acid rain (based on alkalinity values), 19 lakes (22%) are considered moderately sensitive (40-200 μ eq L $^{-1}$ of alkalinity), 52 lakes (61%) as extremely sensitive (0-40 μ eq L $^{-1}$ of alkalinity) while 14 lakes (16%) had 0 or negative alkalinities and, by definition (see page 7), are classified as acidified (Fig. 3). In comparison, the insular Newfoundland survey found 52% of the lakes to be classified as extremely sensitive $(0-40~\mu eq~L^{-1})$, 34% as moderately sensitive $(40-200~\mu eq~L^{-1})$, while 3 lakes demonstrated negative alkalinities. Eight lakes in the island-wide survey were considered relatively non-sensitive and these lakes were essentially restricted to the western and northwestern margin of the province, where carbonate deposits afford unlimited buffering capacity to surface waters. The assessment of the alkalinity values for the south coast confirms the extreme sensitivity of the region's freshwaters, particularly when considering the survey was conducted during the summer season, a period when annual maximums in pH and alkalinity are expected. The fact that the south coast data sets demonstrates greater sensitivity than the island wide data set confirms our concerns that this subregion is extremely susceptible to freshwater acidification. When coupled to the high annual precipitation this region receives and proximity to regional emission sources, this is one area of insular Newfoundland that could be expected to be affected by continuing and/or increasing acidic deposition.

Jefferies et al. (1986), in reviewing a data base of 176 insular Newfoundland lakes, found 61% of the sample to be extremely sensitive ($40~\mu eq~L^{-1}$ alkalinity) and 7% of the lakes to be acidic or acidified ($40~\mu eq~L^{-1}$ alkalinity). In all of eastern Canada, insular NewfounGland was

the second most sensitive region, after Nova Scotia. Jefferies et al. (1986) and others have attributed the extreme sensitivity of lakes in the Atlantic Region, particularly in Nova Scotia and insular Newfoundland, to be related to the widespread occurrence of silicous bedrock overlain by thin soils and/or wetlands.

Measured alkalinity provides an indication of a lake's sensitivity to potential acidification under current depositional regimes. It is, in effect, a measure of residual alkalinity, considering there is evidence consumption of bicarbonate alkalinity in response to acid loadings from both anthropogenic and natural Predictive models (such as Henriksen's Nomograph and Thompson's Cation Denudation Rate model) have been developed from the assumption of stoichiometry between base cations (Ca+1 and Mg and bicarbonate in unstressed waters or as a pre-acid loading condition. Acid loading results in loss of buffering capacity (HCO₃) due to H^T neutralization with concurrent replacement by acid anions (mostly sulphate). Henriksen (1979) suggests that lake acidification can be measured by the decrease in alkalinity over time, with the time span covering the period of pre-acidic deposition to present day. In the absence of alkalinity data prior to the acidification of precipitation, original or pre-acidification alkalinity can be estimated assuming that chemical weathering of minerals by carbonic acid yields equivalent concentrations of cations and HCO, ... Original alkalinity is therefore estimated from current concentrations of cations, most frequently calcium and magnesium.

consideration of cation-alkalinity proportionality depends on several assumptions. The first is that the rate of cationic weathering not increased by strong acid deposition. Several researchers (e.g. Dickson 1975, Gjessing et al. 1976, Dillon et al. 1979) reported increased weathering rates in regions subject to increasing acidic precipitation, and this subsequently led Henriksen (1982) to propose a base cation increase factor for use in empirical predictions of acidification (although he states the data did not clearly resolve the issue of increases in base cation concentrations relation to acid inputs). Researchers now believe that the rate of weathering of base cations has increased in high deposition areas (Schindler 1988). In regions of high deposition, the determination of alkalinity deficit by computing "excess" calcium and magnesium minus alkalinity can lead to overestimates of original buffering capacity and hence the amount of alkalinity loss.

In addition, when alkalinity is determined in the laboratory, bases other than HCO₃ may be titrated, and the relative importance of these species increases as HCO₃ concentration declines (Stumm and Morgan 1981). These other bases can include weak acids (particularly humics and fulvics), ammonia, metal oxides (notably aluminum and iron oxides), and silicates (Kramer 1976). Due to preponderance of coloured waters in Newfoundland, there has been considerable attention paid to the role of organics in acidification and as a potential buffer in acidic, low alkalinity waters. An evaluation of Gran plots from alkalinity titrations for highly coloured and clear bicarbonate poor lakes did not

reveal any quantitative contribution of organic ions in the buffering regime over the range in pH of 3.5 to 5.5. Clair (1987), however, has demonstrated the important buffering role of weak organic anions in low pH (5.5), bicarbonate poor, waters in Nova Scotia.

Assuming base cation concentrations to represent pre-acid deposition alkalinity, the non-marine concentration of calcium and magnesium minus measured alkalinity gives us an estimate of alkalinity loss (deficit). A plot of excess calcium and magnesium versus alkalinity therefore gives us a graphic presentation of the extent and magnitude of alkalinity depletion in south coast lakes (Fig. 6). The deficit ranges from 0.8 to 119.7 μ eq L⁻¹ (\vec{x} = 33.4), with 47 lakes (57%) having deficits in the 20-50 μ eq L⁻¹ range while two lakes demonstrated surplus alkalinity. The alkalinity deficits for the island-wide survey were also mostly in the range of 25 to 50 μ eq L (Scruton 1983) while insular Newfoundland rivers 1986) demonstrated annual deficit in alkalinity greater mean $(\vec{x} = 68 \ \mu \text{eg L}^{-1})$, owing to considerations of seasonal variability in buffering capacity. In 57 (68%) of the south coast lakes, the deficit in alkalinity is greater than the residual alkalinity.

These data provide evidence for the initial effects from acidification, that is, appreciable loss of buffer capacity without widespread chronic pH depression and total loss of acid neutralizing capacity. Fourteen lakes demonstrated negative alkalinities suggesting a component of the data set is demonstrating acidification from natural or anthropogenic sources. While the majority of dilute lakes with appreciable alkalinity deficits are not demonstrating chronic acidification, they may be particularly susceptible to episodic effects in response to heavy fall rains or spring snowmelt. Due to the low residual alkalinity values, small increases in (H) acid loading could greatly increase the proportion of south coast lakes with exhausted alkalinity (previously defined as acidified by the authors).

Howell and Brooksbank (1987) have reported an alkalinity deficit (loss) in 34 Environment Canada (IWD) LRTAP lakes in insular Newfoundland of from 20 to 70 μ eq L (averaging 30 μ eq L) providing evidence of mineral acid loading in addition to background sulphate. Alkalinity losses observed in south coast basins of Newfoundland were partially attributed to the effects of LRTAP by the authors.

CONDUCTIVITY

Conductivity provides a measure of the total concentration of charged ions in a water sample and, as such, is an indication of the degree of mineralization of the sample, but does not necessarily indicate the relative quantities of the various ions. Conductivity has been employed by some researchers as a convenient indicator of sensitivity to acidification (Harvey et al. 1981). Freshwaters with conductivities of 30 $\mu {\rm S~cm^{-1}}$ or less are considered critically sensitive to the effects of acid deposition, and this would include 85 (96%) of the south coast lakes. Comparatively, 86% of the island-wide lake sample and 96% of Labrador lakes were considered critically

sensitive on the basis of specific conductance (Scruton 1983, 1984). Howell (1986) has reported conductivity in Environment Canada's, (IWD) LRTAP monitoring lakes in Newfoundland to range from 14 to $56~\mu \rm Scm^{-1}$ with lowest conductivities observed in Gros Morne and south coast lakes. Other factors, however, such as the influence of the marine environment or the specific activities of each ion (the high activity of H as an example), can limit the usefulness of conductivity as an indicator of acid sensitivity (Haines and Akielaszek 1983).

Conductivity is commonly related to the lithology of a drainage basin. In this data set conductivity was positively correlated to the principal products of geochemical weathering, calcium (r = 0.19, P < 0.05, n = 89), magnesium (r = 0.21, P < 0.051, n = 87), and bicarbonate (r = 0.22, P < 0.05, n = 75). In a region such as the south coast with strong marine influence on freshwater chemistry, ions of marine origin can obscure geochemical sources contributing to conductivity, particularly in the more dilute waters. The marine influence on conductivity was apparent in a positive correlation with chloride (r = 0.26, p < 0.05, n = 89) and the fact that non-marine concentrations of calcium and magnesium were more poorly correlated with conductivity than were total concentrations.

ORGANIC INFLUENCES

The distribution of lakes by colour values compares favorably with the previous island-wide lake survey. The majority of insular Newfoundland's freshwaters are brown water systems or highly coloured lakes (89% in this survey as compared to 84% in the island-wide survey) reflecting moderate to strong influences of natural organic acidity on these systems. It is readily apparent this organic influence is widespread and not an isolated phenomenon. The 34 Environment Canada (IWD) LRTAP lakes demonstrated colour values of 5 to 140 hazen units and only one lake had a value in excess of 100 (Howell 1986). In these lakes water colour was correlated (r = 0.76, P \(\) 0.01) with dissolved organic carbon (DOC).

In characterizing the clearwater study lakes (n=10), 7 were true headwater lakes, 6 were located in the southwest corner of the island (west of Burgeo), and 7 were located on granitic bedrock. Watershed area to lake area ratios (WA:LA) ranged from 3.3 to 16.6 ($\overline{x}=8.2$), considerably less than the sample average (13.7). Kerekes (1978) found that clearwater lakes in insular Newfoundland are usually headwater lakes with low relative basin size (WA:LA ratio), with the likelihood of organic deposits within a watershed being related to this ratio. In addition, clearwater lakes are common in areas of the Long Range Mountains due to the absence of organic deposits at higher elevations, owing to high watershed slopes and rapid runoff. Six of the ten clearwater lakes were located in the Long Range and Annieopsquotch Mountains, at elevations ranging from 251.5 to 468.4 m above sea level. The relationship between lake drainage order and the occurrence of clearwater lakes is largely related to the frequency of occurrence of organic deposits; that is to say the higher the lake order, the more individual lake drainages feeding into that lake and consequently the greater the likelihood of contribution of coloured waters from upstream lakes.

Wells and Pollett (1983) have characterized the peatlands in the study area as being largely of basin bog type (to the south, within 25 km of the coast) and slope bog type (to the interior in the headwaters of the major river drainages) with some Atlantic ribbed fen (below and to the east of Meelpaeg Reservoir). These bog types are characterized by smaller peat deposits, operational at a watershed scale, as opposed to blanket and plateau bogs which dominate broad geographical expanses. Seepage waters are characteristically acidic (pH of 3.5 to 4.0) and nutrient poor. In this type of bog development, a highly coloured headwater lake and a clearwater lake can have adjacent watersheds. Not surprisingly, there is no apparent spatial trend the distribution of lakes classified as brown water and highly coloured systems, but 8 of 10 clearwater lakes were located in the western half of the south coastal region (Fig. 7).

The considerable influence of organics on freshwater lake chemistry in insular Newfoundland and Labrador has been previously demonstrated (Earle et al. 1986, 1987). Factor analysis of extensive data sets for insular Newfoundland (n = 109, Scruton 1983) and Labrador (n = 130, Scruton 1984), revealed that two principal Scruton 1984), revealed that two principal factors, hardness and dystrophy, accounted for greater than 50% of the total variance in chemistry in both data sets. For the insular Newfoundland data set, dystrophy was the most important factor, accounting for 27.5% of the variance. The dystrophy factor was positively correlated (P < 0.10) to colour (r = 0.91), iron (r = 0.82), aluminum (r = 0.81), carbon dioxide (r=0.68), turbidity (r=0.65), manganese (r=0.64) and sulphate (r=0.40) while the factor was also negatively correlated to pH (r = -0.36) and secchi depth (r = -0.72). The factor analysis revealed that, on a broad-scale regional basis, bedrock/surficial geology and the widespread occurrence of organic deposits (as soils or in peatlands) are the principal determinants of freshwater chemistry. A salinity or marine influence factor was deemed to account for 18% and 10% of the variance in insular Newfoundland and Labrador, respectively. It is noteworthy that a LRTAP factor, reflecting the influence of anthropogenic acidification, did not emerge in either N.I.S. data set.

An important question to emerge in recent years is whether the occurrence of natural organic acidity can explain the observed lake acidification (loss of acid neutralizing capacity, declines in pH) in Eastern Canada, and how important weak acids of natural origin are in relation to strong acid deposition in precipitation. Organic acids are extremely important in dilute surface waters for two reasons: they contribute significantly to acidity and they chelate available metals thereby increasing total solubility and decreasing biological toxicity (Dickson 1980; Oliver et al. 1983; Lazerte and Dillon 1984; Jefferies et al. 1986; Rosenquist et al. 1980). In addition, weak organic anions, may play an important role in the

buffering regimes of bicarbonate poor, coloured lakes.

Jefferies et al. (1986) have reviewed the organic content of over 800 lakes in Eastern Canada, including 253 lakes on the island of Newfoundland. The median organic anion concentration for Newfoundland lakes was 56 μ eq L 1 COOH, while 19% of the sample exceeded 100 μ eq L 1 . These values compared favourably with COOH data collected in this survey (x COOH = 41.4 μ eq L 1 , 6% exceeding 100 μ eq L 1). The insular Newfoundland sample (n = 253) had the highest median organic content of all 8 eastern Canadian subregions with the exception of Northwest Ontario (median COOH of 94 μ eq L 1 , 42% \geq 100 μ eq L 1 , n = 47). Due to the relatively low acid neutralizing capacity of south coast lakes, and relatively low anthropognic sulphate component, organic acids have become a dominant anionic component (Table 2).

Jefferies et al. (1986) also reviewed the interrelationships between acid neutralizing capacity (ANC) and organic anion concentration (COOH) in Eastern Canada. On a broad geographical basis, natural sources of organic acidity were not considered to be the predominating influence on lake acidity in freshwater lakes in eastern Canada. This situation was true for most of the subregional data sets, particularly in northeastern Ontario where acidic lakes had low organic anion concentrations, and hence were considered classical clearwater acid lakes. In this data set, alkalinity (or ANC) was unrelated to DOC or organic anion concentration (COOH).

Howell (1986) has used the residual of MTB-sulphate minus IC-sulphate as an estimate of organic anions in 34 Environment Canada (IWD) LRTAP monitoring lakes, as opposed to the method of estimation of Oliver et al. (1983), as employed in this study. Organic anions, as estimated the residual of the sulphate measures, varied from 0 to 40% of the total anions while the estimate obtained by the Oliver et al. (1983) method found the organic component of the anions to vary from 6 to 89%. The authors felt the Oliver et al. method likely estimates 'potential' organic anions, likely estimates 'potential' organic anions, including both true ionic and complexed forms. Howell and Brooksbank (1987) have suggested that variability in relationships between water colour and quantitative organic content (DOC) through drainage basins, regions, and seasons in Atlantic probably indicates that the different component acids (species) vary both spatially and temporally. This is an important factor to consider when quantifying organic acid concentration as the general reactive radical COOH (as Oliver et al. 1983 has).

Patrick et al. (1981) have suggested that acidification of naturally acid dystrophic systems (pH \(\frac{5}{2}.25\)) may not seriously affect resident aquatic biota. The authors maintain that naturally acid lakes generally contain low diversity of acid-adapted fauna that would be largely unaffected by a shift to lower pH regimes. In addition, the presence of particulate or dissolved humic material results in chelation of heavy metals (e.g. aluminum, manganese) liberated from lake catchments by strong acid deposition,

rendering them non-toxic to resident fauna. However, Davis et al. (1985) have found evidence in the paleolimnological record of Norwegian lakes to suggest that, in a region with a long-standing (several decades) record of high levels of atmospheric acid loading, weak organic acid dominated systems can shift to become strong mineral acid dominated.

SULPHATE

Sulphate values for south coast lakes were lower than those previously recorded during the island-wide survey of 1981. This is somewhat surprising as sulphate values on the south coast were expected to approach the upper range for the island, owing to regional patterns of precipitation and assumed anthropogenic deposition. Comparing sulphate data for the 24 south coast lakes surveyed in 1981 (excess sulphate of 20.9 to 59.8 μ eq L $^{-1}$, \overline{x} = 32.8) with the 1983 data (5.4 to 44.4 μ eq L $^{-1}$, \overline{x} = 21.0), levels in 1983 are appreciably lower, possibly as a consequence of analytical differences. This may also reflect a lower rate of acid loading in 1983, which is consistent with trends in North American sulphur emissions (Martin and Brydges 1986).

Seasonal variability may also play a role in the low excess sulphate values recorded for south coast lakes. While sulphate does move rather freely through aquatic systems and, on the long term, input-output budgets are rather constant, there is a marked seasonal flux in movement through a watershed linked to the hydrological (Skartveit 1981). Sulphate generally cvcle accumulates during the low flow periods, largely through vegetational uptake during the period of plant growth in summer months, and through snow ; pack accumulation in the winter (Dovland and Semb 1978). A net loss in sulphate (output exceeds input) as a result of washout during high flow periods is often characteristic of fall months (heavy rains) and the spring snowmelt period. Organic soils, which predominate in the south coast drainages, also can act as biogenic sulphur reservoirs and experience flushing during hydrological peaks. As previously discussed, a review of hydrometric data for gauged rivers within the study area indicated the 5 to 6 weeks prior to the survey were characterized by annual lows in the hydrological cycle, followed by a small 2-day spate. The fact that this survey was conducted at/or near the low point in the hydrological cycle during the growing season may in part, explain, the relatively low sulphate values recorded in this study. Kerekes and Howell (1983) and have found the lowest seasonal sulphate values in Nova Scotia (Kejimikujik National Park) streams to occur during the summer growing season.

In the previous island-wide survey, geological sources of sulphate were suspected in 4 of 109 lakes (Scruton 1983). High excess sulphate values that occurred in association with elevated iron values were evaluated as evidence of pyrite or gypsum deposits within the watersheds. This excess sulphate/iron relationship suggested pyrite influence on lake chemistry in central Newfoundland (northeast of Meelpaeg Reservoir) and on the western margin (Flat Bay-Lower Long Range Mountains) of the island. Excess sulphate values (non-marine) in these lakes were considerably

higher than in other systems and ranged from 70.9 to 89.2 μ eq L⁻¹. Sulphate values recorded for south coast lakes in 1983 were much lower than these levels and, although iron was not measured in this survey, geological inputs of sulphate were not suspected. Pyritic formations are known to occur in association with gold deposits on the south coast, particularly in the Cinq Cerf River watershed and the upper Isle aux Morts River watershed. These deposits are small and localized and do not appear to have influenced the chemistry of lakes sampled within those river drainages.

this study was determined (MTB method) and by ion Sulphate in this determined colourimetrically chromatography (IC). Humic substances in colored waters are known to interfere with colourimetric determinations leading to an overestimate of sulphate concentrations (Cronan 1978). As previously discussed, MTB sulphate was highly correlated to IC sulphate and the difference (residual) between the two determinations (MTB.SO₄ ¹-IC.SO₄ ²) was significantly correlated to water colour, providing strong evidence of colourimetric interference. Underwood et al. (1983) examined data from lakes in Nova Scotia and Newfoundland (with colour values in the range of 50 to 100 TCU) and found the error in sulphate determination (overestimation by the MTB method) to range from about 12 to 33 μ eq L⁻¹. Kerekes (1983) cautioned against using any MTB/IC sulphate relationship to correct historical data for colorimetric interference. He found the colour versus (MTB-IC) SO₄ relationship to vary seasonally as well as geographically, and the relationship was not always linear. Sulphate determination in acid rain studies in Newfoundland is now routinely completed by ion chromatography. In the absence of an ion chromatograph (pre 1982-83), sulphate had been routinely determined by the MTB method after a pre-treatment by ultraviolet irradiation, to minimize the colourimetric biases in SO₄ values. It is colourimetric biases in SO₂ values. It is recognized that most historical sulphate data (almost exclusively determined by MTB) in Newfoundland, including the 1981 island-wide survey, contains biases (overestimation) due to the preponderance of humic waters in the region. This complicates comparisons with historical data and assessment of acidification trends.

Non-marine sulphate was strongly positively correlated to the two principal ions arising from marine deposition, Na⁺ and Cl (r = 0.60 for both, P \leq 0.001), and negatively to distance from the coast (r = -0.42, P \leq 0.001), suggesting possibly insufficient sea salt correction. In addition to sea-salt inputs, and possible geochemical sources of sulphate, it is acknowledged there may be natural (background) levels of sulphate in lake water, largely of biological origin. Harvey et al. (1981) indicated background sulphate levels in Canadian Shield lakes could be as much as 30-60 μ eq L⁻¹, while Almer et al. (1974) suggested sulphate background levels from 20 peq L¹ to 60 peq L¹ for Swedish lakes. Recently, Galloway (1986) has suggested a global background level of approximately 5 μ eq L⁻¹ of SO, in precipitation, arising from non-anthropogenic sources. Work on the Kejimikujik calibrated watershed in southwestern Nova Scotia found correlations between organic carbon and sulphate, indicating that organic deposits (and soils) ray act as sulphur reservoirs

by storing reduced sulphur compounds (Kerekes et al. 1982). Bacterial action produces sulphate from these reservoirs which can be transported to receiving waters during periods of high runoff. In peatlands, sulphate is taken up deep in bogs during dry periods (water table is reduced) and can be subsequently released (Urban and Bayley 1986). Excess sulphate has been assumed to be a reliable surrogate of the magnitude of anthropogenic deposition, but it is apparent there are other possible sources, as well as seasonally operative processes, which complicate the scenario.

Howell and Brooksbank (1987) have made an important observation in reviewing lake sulphate data from insular Newfoundland. In their data base, excess sulphate concentrations (determined by IC) in highly coloured Newfoundland lakes were considerably lower than values observed from clearwater lakes within the same geographical area, suggesting that highly coloured lakes in Newfoundland may be either storing or exporting elemental or reduced sulphur compounds. The authors suggest that as much as 50% of the sulphur in some Newfoundland lakes may be in the reduced or organic forms. This further complicates the use of (excess) sulphate as a surrogate for hydrogen ion loading from LRTAP sources, particularly in the organic dominated systems.

Another important consideration in evaluating lake sulphate data is the temporal pattern in sulphate deposition, both seasonally (as discussed) and over the long term (years). It is apparent that SO emissions in North America have demonstrated a downward trend from the 1970 to 1973 period to the early 1980's (1980 to 1983), with 1983 emissions being the lowest over the preceding decade (Fig. 8). Thompson and Bennett (1981) have demonstrated an inverse relationship between sulphate concentrations and pH in the Rocky River in Newfoundland which also appears to reflect the reduction in sulphate emissions since 1973. This pattern of declining sulphate emissions (and presumably deposition patterns) to 1983 may be reflected in the lake chemistry. South coast lakes are mostly small and shallow with steep watershed slopes. Due to the high annual precipitation this region receives, these lakes are expected to flush rapidly, in the order of a few days to weeks. Small ponds in Central Newfoundland, a region of lower annual precipitation and low watershed slopes, flush from 6 to 50 times per year (Ryan and Wakeham 1984). Due to the rapid flushing of these south coast lakes, water chemistry will reflect both the residual buffering capacity in the watershed (pH, alkalinity, base cations) and the recent magnitude of acid loadings (H, SO4 '').

Data from the APN (Air and Precipitation Monitoring Network) station at Baie D'Espoir on the south coast (for the seven months preceding the survey, December 1982 to June 1983) was reviewed to provide insight into recent sulphate loading to south coast lakes. This network ceased operation in mid-1983 (July) and became part of the CAPMoN network (Canadian Air and Precipitation Monitoring Network). The weighted mean rainfall pH (monthly) varied from 4.69 (May 1983) to 5.25 (January 1983) (weighted mean of 5.09) while total wet sulphate in precipitation varied from 10.5 to 44.0 $\mu{\rm eq}~{\rm L}^{-1}$ (weighted mean of 20.1). Rainfall

acidity for the seven months preceding the survey was less than the previous reported range in rainfall acidity for the province of pH 4.5 to 4.9 (Anonymous 1983). The downturn in continental sulphate emissions, particularly in relation to the closure of smelters in Ontario/Quebec in 1983, may have resulted in lower acid loadings in early 1983 in southern Newfoundland, which may also be reflected in lake sulphate concentrations.

Howell and Brooksbank (1987) have reported excess sulphate levels in southwest Newfoundland lakes to be slightly higher than those recorded from lakes in Cape Breton Island in Nova Scotia. Assuming lake excess sulphate to represent acid loadings, this observation is somewhat inconsistent with the west to east gradient in acid deposition in Atlantic Canada. Higher values on the south coast could be related to different air masses affecting the two regions, but may also be evidence of the influence of sulphur emissions from the Lingan Generating Station on the south coast of Newfoundland.

Lake sulphate values demonstrated no relationship with elevation (sulphate vs. elevation, $r=0.01,\ P=0.90;$ excess sulphate vs. elevation, $r=0.02,\ P=0.87)$. It has been hypothesized that high elevation lakes experience higher rates of deposition (precipitation) and thus should exhibit greater sulphate values than lower elevation lakes, within the same region (Kelso et al. 1986). Sulphate on the south coast (both total and non-marine) did exhibit a gradient with respect to distance from the coast but not in relation to topography. The relatively narrow range in both lake sulphate values and lake elevation likely contributes to the absence of any correlation. In the island-wide survey, total sulphate was correlated (r = -0.27, P \leq 0.005, n = 109) to lake elevation but non-marine sulphate was not. These relationships suggest that deposition of the marine component of sulphate is influenced by considerations of topography and proximity to salt water, while deposition of the (presumed) anthropogenic component may unrelated to elevation.

Oden (1976) has proposed that the ratio of bicarbonate alkalinity to excess sulphate be used as a preliminary indicator of progressing acidification. Geochemical weathering in regions influenced by acidic precipitation is accomplished by both carbonic acid and strong acids (mostly sulphuric and, to a lesser extent, nitric). As precipitation acidity increases, sulphuric acid plays an increasing role in the weathering process, resulting in the gradual replacement of bicarbonate by sulphate in receiving waters. In low alkalinity waters, sulphate eventually replaces bicarbonate as the dominant (non-marine) anion, resulting in HCO₃:SO₄ ratios of less than 1.0

A plot of the HCO_3 /Ex.SO $_4$ 7 ratio versus lake pH (Fig. 9) reveals that sulphate is dominant to bicarbonate in 42 lakes (50%), with these lakes having pH's ranging from 4.61 to 6.60, close to the range in acidity for the entire sample. Of these, 28 lakes had ratios of less than 0.5 indicating twice as much sulphate as bicarbonate, and this includes the 14 lakes with no titratable bicarbonate. Owing to the relative narrow range in both alkalinity (0 to 188.5 ρ eq L $_1$ 1 with only

three values exceeding 100.0) and non-marine sulphate (5.4 to 44.4 μ eq L⁻¹), it is difficult to ascribe the trend in the HCO₃/Ex.SO₄⁻² ratio to variation in either parameter. Generally, 78% of lakes below pH 5.80 are sulphate dominated (ratio of (1.0) while above pH 5.80, 80% of the lakes were bicarbonate dominated (ratio >1.0). Comparatively, 44% of the 109 lakes in the island-wide survey were sulphate dominated, with this dominance particularly evident in lakes less than pH 6.30. In this study, it is apparent that, despite relatively low values (at the time of sampling), sulphate is becoming the dominant anion relative to bicarbonate in acidic, low alkalinity lakes. However, considering all the non-marine anions, organics are frequently the most important anion (in quantitative terms) in lakes of pH 5.3 and less. The order of ionic dominance may also be confounded by the uptake of SO₄⁻² in bogs and the seasonality of this process (Urban and Bayley 1986).

The geographic distribution of the bicarbonate to excess sulphate ratio (HCO₃/Ex.SO₄²) indicates that values of less than 1.0 are distributed across the study area, with a higher proportion of excess sulphate dominated lakes in the southwest corner of the province (Fig. 9). This is largely a consequence of the occurrence of both higher lake sulphate and lower alkalinity values in the same region (Fig. 5 and 7). However, on the western slope and plateau region of the Long Range Mountains, several lakes (#600, 606, 608, 611, 613, 632) have ratios greater than 2.0, despite relatively higher sulphate values. The previous island-wide survey had found inconsistencies between lake chemistry and bedrock geology along the western slope of the Long Range Mountains, which was attributed to localized patterns of glacial redistribution of overburden (with the overburden having relatively higher buffering potential). This may also be the situation for the above six lakes.

ALUMINUM

Total aluminum (Al) values for the south coast study lakes were found to have a wider range and greater mean (33 to 554 μ g L $^{-1}$, \overline{x} = 357) than those for the island-wide survey of 1981 (0 to 430 μ g L $^{-1}$, \overline{x} = 110). Most values (88%) in the previous island-wide survey were less than 200 μ g L $^{-1}$, while in this study aluminum exceeded this value in 83 lakes (92%). In the 1981 survey, aluminum had been negatively correlated with lake pH (r = -0.48, p < 0.001) and positively related to water colour (r = 0.81, p < 0.001), while these relationships are not apparent in the south coast data. Howell (1986) reported aluminum concentrations of from 20 to 300 μ g L $^{-1}$ for 34 Environment Canada (IWD) LRTAP lakes, with aluminum content exceeding 200 μ g L $^{-1}$ in only 9% of the samples. In the IWD lakes, aluminum content was strongly correlated to water colour (r = 0.62, p > 0.01, n = 214). There are no apparent explanations for the absence of aluminum-pH or aluminum-colour relationships in the south coast data, or for the appreciably higher aluminum values recorded.

Organic ligands (presumably a correlate to water colour) enhance aluminum solubility by forming organic-Al complexes which are transported

through soil profiles into receiving waters (Johnson 1979). Consequently, organic aluminum fractions frequently are the dominant species of the metal available in coloured waters resulting in strong colour-Al relationships (Stumm and Morgan 1981). Clair and Komadina (1984) found about 95% of the aluminum in Nova Scotian freshwaters (principally Kegimikujik National Park) to be in the organically complexed form. Additionally, aluminum demonstrates increasing solubility with declining pH (over the 7.0 to 5.0 As soil acidity increases due to ic deposition, humification, etc. range). atmospheric deposition, humification, etc. aluminum can be released into surface waters yielding negative correlations between lake water pH and aluminum.

In previous studies, the strong relationships between aluminum and water colour had suggested that much of the available ion in receiving waters is organically complexed (Scruton 1983, 1984). Organic complexes of aluminum are relatively non-toxic to aquatic biota, as compared to monomeric forms, and consequently it was assumed that little toxic aluminum is present in Newfoundland waters. The absence of such a relationship in south coast lakes, however, does not allow us to draw similar conclusions with respect to aluminum speciation and toxicity.

Aluminum in south coast lakes also appears to be above levels reported for lakes of intermediate pH (5.4 to 6.0) in Norway and Ontario (Al of 50 to 90 $\mu g \ L^{-1}$) and in Finland (Al of 80 to 100 $\mu g \ L^{-1}$) (Harvey et al. 1981, Anonymous 1982, respectively). Acid affected lakes in Scandinavia and North America have reported a range in aluminum values from 200 to 600 $\mu g \ L^{-1}$ (Dickson 1975 for Swedish lakes), 50 to 300 $\mu g \ L^{-1}$ (Wright and Henriksen 1978 for Norwegian lakes), 80 to 700 $\mu g \ L^{-1}$ (Spry et al. 1981 for Ontario lakes), and 20 to 510 $\mu g \ L^{-1}$ (Bobée et al. 1982 for Quebec Lakes). The comparison of lake aluminum data from south coast lakes with data from acidic and intermediate pH lakes, as well as with available data for insular Newfoundland, does suggest an inherent bias (overestimation) in the data set. Quality assurance checks did indicate a slightly high bias (overestimation) in aluminium determination but the bias was not considered great enough to invalidate the data.

CALCITE SATURATION INDEX

The calcite saturation index (CSI) has been adopted as a reliable indicator of sensitivity to acidification (Kramer 1976) in that incorporates both geology and water chemistry into a convenient reference index. The CSI of a water sample is the logarithm of its degree of saturation with respect to calcium carbonate. Water supersaturated with calcite will have a CSI greater than zero, while undersaturated water will have a CSI of less than zero (negative values). It has been proposed that a CSI value from -2 to -3 indicates potential danger from acidification while values of less than -3 indicates extreme potential for acidification with deleterious effects likely (Gorham 1976). Bobée et al. (1982) have considered lakes with CSI values of between -3 and -5 to be 'potentially vulnerable', while lakes with values of less than -5 would be considered 'extremely sensitive'.

The CSI for the study lakes ranged from -3.60to -7.87. Five (7%) lakes had CSI values of -3 to -4, 23 lakes (33%) from -4 to -5, 20 lakes (29%) from -5 to -6, 17 lakes (24%) from -6 to -7 and 5lakes (7%) had CSI values of -7 or less. The CSI was not calculated for the 14 lakes with zero or negative alkalinities as these systems have no measurable bicarbonate. These values are consistent with the absence of calcareous deposits throughout southern Newfoundland, and the low buffering capacity of region's freshwaters. Clair et al. (1982) used historical water quality data to conduct contouring analyses of CSI values for the island of Newfoundland. The results indicated the south coast region was characterized by CSI values of $\sim\!4.0$ and less, which is largely consistent with inventory data collected in 1983. The spatial distribution of CSI values is displayed in Fig. 10.

Lake inventory data from Fisheries and Oceans' NIS Survey (n = 814) indicated that the majority of the lakes surveyed in Eastern Canada (77%) had CSI values of less than -3 and were therefore classified as sensitive to acidification (Kelso et al. 1986). The proportion of sensitive lakes by region, based on CSI values, was as follows: Nova Scotia (95%), Labrador (92%), insular Newfoundland (87%), Quebec (73%), New Brunswick (72%) and Ontario (62%). The south coast lakes, all with CSI values of less than -3.0, represent a broad region whose freshwaters are characterized as homogeneous with respect to their acid sensitivity. Sixty percent of the lakes had CSI values of less than -5.0 and could be classified as extremely sensitive (after Bobée et al. 1982).

pH-CALCIUM RELATIONSHIPS

Henriksen (1979) has used considerations of base-cation to alkalinity equivalency in pre-acid stressed waters to develop a simple approach to identifying acidification. His original model, called an 'Indicator Diagram', is based on the premise that pre-acidification alkalinity, and hence pH, can be estimated from current non-marine calcium values. The model derives from the assumption that in dilute oligotrophic waters, alkalinity is proportional to the sum of calcium (Ca⁻¹) and magnesium (Mg⁻¹), the ratio of Ca⁻¹ to Mg+1 is generally constant, and that alkalinity is function of pH. His model employs empirically developed curve to separate 'acidified' and 'non-acidified' freshwaters, considering that acidification in this sense is defined as significant loss in alkalinity but not necessarily total alkalinity depletion and chronic pH depression. Under this definition, the model is able to identify lakes with apparently safe ph's that may have been affected by appreciable losses in alkalinity.

The pH-excess calcium relationship for south coast lakes is plotted with Henriksen's empirical curve in Fig. 11. Seventeen (20%) of the lakes fell within the 'acidified portion' of the figure, while the remaining lakes fell below the empirical curve. These 17 lakes fell largely into two geographical clusters. Six lakes were located in the southwest corner of the island (%'s 602, 603, 614, 615, 617, and 628) in five different river

drainages, while an additional eight of lakes (#'s 661, 662, 669, 670, 671, 672, 686, and 687) were located in the central sub-region, with five of these lakes located in the expansive headwaters of the Salmon River drainage. These lakes are generally characterized by low pH's (4.61 to 5.74, $\overline{x}=5.09$, with 94% less than pH 5.50) and extremely low alkalinity values (-29.6 to 25.2 $\mu eq~L^{-1}$, $\overline{x}=2.5$). Alkalinity deficits in these lakes ranged from 23.3 to 67.0 $\mu eq~L^{-1}$ ($\overline{x}=41.4$), which is somewhat higher than the mean deficit for the entire data set (33.4 $\mu eq~L^{-1}$).

Excess sulphate values for these 'acidified' lakes is similar to that for the entire data set $(\overline{x} = 22.2$ and $21.0~ \mu eq~ L^{-1}$, respectively), consequently atmospheric loading cannot be identified as the principal cause for alkalinity These lakes are all brown water and depletion. These lakes are all brown water and highly coloured systems (TCU values from 35 to 100, $\overline{x}=75$) reflecting considerable organic influence on chemistry. In 13 of these lakes, organics (COOH) are the dominant anionic constituent. The 8 lakes in the central cluster were organic dominated, while 3 of the 6 lakes in the southwest corner were (non-marine) sulphate dominated. High annual precipitation and runoff levels in the southwest portion of the south coast may contribute to higher acid loading rates in this region, which lake sulphate concentrations do not represent (mass balance considerations). The rate of acid loading may be more accurate represented by sulphate dominance and alkalinity deficits, but is not necessarily reflected in the modest lake sulphate concentrations. It is also noteworthy that these 'acidified' lakes were mostly small (15 or 88% were less than 30 ha), while the two larger lakes (100, 238 ha) were headwater systems with very low watershed area to lake area ratios (less than 2.5). These lakes, therefore, possess hydrological characteristics that contribute to their susceptibility.

HENRIKSEN'S NOMOGRAPH

Henriksen (1980) has developed a model (or nomograph) to evaluate lake chemistry data in relation to stages in the process of freshwater acidification, considering that the process is continuous and dynamic. The underlying premise of the model is that the process of acidification is analogous to the titration of a bicarbonate buffering system (of geochemical origin) with strong mineral acids (of atmospheric origin), more specifically with dilute sulphuric acid. model also derives from the basis of equivalency between the major cations (calcium and magnesium), or calcium alone, to bicarbonate in non-acidified waters, and the use of present cationic concentrations to estimate original (pre-acid loading) alkalinity. Non-marine calcium and magnesium is plotted against non-marine sulphate such that the ordinate represents geological determinants and the abcissa represents anthropogenic inputs. The nomograph was developed from empirical data for 719 small southern Norwegian lakes, and was subsequently tested in relation to other data sets. The model is divided into three sections corresponding to segments in the titration of a bicarbonate buffered system. The first stage (Bicarbonate) is characterized by small decreases in alkalinity but maintenance of an intact, bicarbonate dominated, buffering system. The second stage (Transition) is characterized by the loss of the buffering regime over time, severe fluctuations in pH, and episodic effects on biota. The final stage (Acid) is characterized by chronically low pH's, elevated levels of trace metals, and severe perturbation of resident biota.

Prior to evaluating data from south coast lakes on the nomograph it is important to review the assumptions implicit in model development and application. Henriksen (1980, 1982) stated explicable to surface waters with significant concentrations of organic acids, largely because organics can be a major determinant of water pH, independent of atmospheric acid loading. In addition, laboratory analyses of cations in coloured waters include organic complexes of calcium and magnesium, leading to an overestimation of true ionic concentrations.

The model was originally developed assuming that no increase in base cations occurred in response to increased acid loading. Other researchers (Almer et al. 1978, Dillon et al. 1979, and others as reviewed in Harvey et al. 1981) suggested that increased rates of weathering do occur in response to increased rates of anthropogenic acid loading. Henriksen (1982) addressed the issue and, although no universal trend was evident, he proposed a maximum "base cation increase factor" for use in the mathematical expression of the empirical model (to follow). If an increase in rate of weathering does occur in regions with high acid loadings, present day concentrations may cationic overestimate original alkalinity, and hence the magnitude of acidification (as change in alkalinity from pre-acid loading to present day).

The applicability of any model, developed with data from one geographical area, to other regions is a concern. Henriksen did apply his model to other geographical locales in a test of his original hypothesis, however, Bobée et al. (1982), Haines and Akeilaszek (1983), and Church and Galloway (1984) found inconsistencies in application of the model to lake data sets from Quebec, New England, and the Adirondacks, These inconsistencies have been respectively. related to regional differences in bedrock geology (and weathering products), weathering rates, non-anthropogenic natural sources of acidity, and relationships between precipitation chemistry and lake chemistry. This has led to suggestions that it may be necessary to develop nomographs (and regression relationships) that are region-specific (Bangay and Riordan 1983).

Data for the study lakes is plotted on Henriksen's nomograph in Fig. 12. Six lakes (7%) fell in the 'Transition' zone of the nomograph, while the remaining 79 lakes (93%) were located in the 'Bicarbonate' zone. The nomograph correctly predicted the pH of 69 lakes (81%) while the model incorrectly positioned 16 lakes (19%) in relation to the pH ranges of the three stages of the nomograph. Of the 16 lakes that did not conform to the model, 13 of the lakes had pH's from 4.72 to 5.20 and should have fallen in the 'Transition' zone while one lake (#687, pH of 4.61) should have been located in the 'Acid' zone. Twelve of these 16 lakes were very highly coloured (50 to 100 TCU)

and it is probable that natural organic acidity is the primary determinant of pH in these lakes.

the six fell in lakes that 'Transition' zone, the pH of four lakes conformed to model predictions while two lakes (#s 607 and 619 with pH of 5.40 and 5.38, respectively) should have fallen outside the bounds of this zone. These six 'Transition' lakes are characterized as small (all less than 100 ha, $\vec{x}=37.4$), with low watershed to lake area ratios (all < 2.0, \overline{x} = 7.9), while five of the lakes were headwater systems. For five of the six lakes, sulphate values were approaching the upper range for the data set (31.8 to 44.4 μ eq L $^{-1}$, \bar{x} = 38.3) and four were (non-marine) sulphate dominated. Lake #644 (colour of 100 TCU, excess sulphate of 17.3 μ eq L⁻¹) fell within the 'Transition' zone but this lake would appear to also be organic acid dominated. Four of the six 'Transition' lakes were located in the southwest corner of the province where annual precipitation is extremely high (1.8 to 2.0 m).

It is apparent that 'Transition' lakes possess hydrologic properties that characterize them as very highly susceptible to anthropogenic acidification. These lakes were small headwater systems with low watershed to lake area ratios and were largely located in the subregion of insular Newfoundland with the highest annual precipitation (high acid loadings). These lakes may have lower flushing rates than small lakes lower in the drainage system, but their terrestrial pool of potential buffering agents would be much less. Sulphate values in these lakes were modest relative to values from acidified regions of the world (range of 41 to 368 μ eq L $^{-1}$, Wright 1983), although values were higher than most other lakes on the south coast. Five of the 'Transition' lakes were moderately coloured (30 to 60 TCU, $\overline{x} = 50$) so organic contributions to the acid-base balance in these lakes is also a possibility.

Howell (1986) has plotted the data from 34 Canada (IWD) LRTAP lakes Environment Newfoundland and found all lakes to fall in the 'Bicarbonate' zone except one, which fell in the 'Transition' zone. The author feels the excess sulphate data for these lakes suggest a rainfall pH of between 4.7 to 5.0, and the position of the lakes on the nomograph indicated that mineral acid loading is not of sufficient magnitude to have caused large pH declines in the lakes. Howell and Brooksbank (1987) reviewed a larger data set from insular Newfoundland (n = 214) and found lakes on the eastern side of the Northern Peninsula (50% of the regional sub-sample) to fall in the 'Transition' range. Conversely, south coast lakes fell predominantly in the 'Bicarbonate' zone (only 2 lakes were in transition). Organic acidity was considered to be important in the pH of south coast systems and the authors concluded that present mineral acid (LRTAP) loading is not sufficient to completely exhaust the bicarbonate buffering system.

As indicated, the model did not correctly predict pH for 16 of the lakes, and the majority of these lakes were extremely dilute, highly coloured, and organic anion (COOH) dominated. For this lake type, the cation/alkalinity/excess sulphate relationships developed by Henriksen do not appear to be adequate in determination of pH,

and dystrophy appears to be the a regulator. As Henriksen (1980, 1982) has stated, the nomograph may not be applicable (without modification) to highly coloured waters for two reasons; weak organic acids play a major role in the acid/base balance of these waters and, in addition, analytical determination of base cations would include cation-organic complexes, therefore, overestimating true ionic concentrations (and hence alkalinity depletion). Another concern in this data set is the importance of sodium as a major cation in many of the south coast lakes. If sodium concentrations are an indication geochemical weathering and concurrent generation of bicarbonate, then the assumption of calcium and magnesium to bicarbonate equivalency as a pre-acid loading condition is invalidated. These concerns and limitations in application of this model, along with the improper placement of several south coast lakes on the nomograph, suggests the model may not be suitable in assessing the process of lake acidification for a majority of the study lakes.

The inability to adhere to several of the underlying assumptions in model application might suggest that evaluation of data from south coast lakes on Henriksen's nomograph is a wasted effort. The data was, however, evaluated in relation to the empirical considerations of Henriksen (1980) to permit comparison with previous regional data sets and with data from other global areas. The poor applicability of the nomograph also points out a major shortcoming of many of the early empirical approaches to understanding the acidification process; that is that organic dominated systems cannot be quantitatively evaluated for acidification effects. Applying the nomograph to clearwater oligotrophic lakes only on the south coast (this study) and for the island as a whole (Scruton 1983) would result in only a small proportion of the lakes meeting this exclusive criteria.

ION RATIOS

Jefferies (1986) has suggested that certain ionic ratios be used to evaluate geographical patterns of acidic deposition and terrain sensitivity (alkalinity and base cation supply). The principal chemical response of sensitive freshwaters to acidic deposition is decreased (or eventually exhausted) acid neutralizing capability (ANC) coupled to increases in lake sulphate and sometimes base cations (Henriksen 1979). Consequently, three ion ratios can be used to evaluate freshwater response to acidic deposition:

- a) alkalinity: excess calcium and magnesium;
- b) excess sulphate: excess calcium and magnesium; and
- c) alkalinity: excess sulphate.

Alkalinity: Excess Calcium and Magnesium

Bicarbonate alkalinity and base cation levels in water are considered to reflect primarily the process of geochemical weathering and consequently anion/cation concentrations resulting from this process should be proportional. The $Alk:(Ex.Ca^{+\frac{1}{2}}+Ex.Mg^{+\frac{1}{2}})$ ratio should therefore

vary from unity (1.0) in an ideal, clearwater, low deposition lake to zero (0.0) as alkalinity is exhausted, to negative values as the lake becomes increasingly acidic. Jefferies (1986) has demonstrated that this ratio is sensitive to the absolute value of the numerator (Alk.) and denominator (Ex.Ca¹ + Ex.Mg²); that is to say the ratio will decline to a lesser amount in lakes with high alkalinity and base cation levels than in lakes with lower concentrations for these ions, in response to acid loading. Hence, the magnitude of the response of this ratio to acid loadings directly reflects the ability of the terrain to supply base cations and alkalinity.

The spatial distribution of the Alk:(Ex.Ca $^{+2}$ + Ex.Mg $^{+2}$) ratio for the southcoast lakes is shown in Fig. 13. Eleven lakes (13%) had negative ratios, 17 lakes (20%) had ratios in the 0 to 0.20 range, 32 lakes (38%) in the 0.21 to 0.50 range, and 25 lakes (29%) had ratios from 0.51 to 1.00. Broadscale contour maps presented in Jefferies (1986), using a data base of 268 lakes for insular Newfoundland, had previously demonstrated two areas of the island to have ratios less than 0.2, the aand from the Central Highlands of Newfoundland to south coast and the eastern side of the Northern Peninsula. There is no obvious trend to spatial distribution of the Alk:(Ex.Ca⁺ + Ex.Mg⁺) ratio with negative values (theoretically acidified) and low ratios (0.00 to 0.20) being distributed across the south coast. A greater proportion of higher ratio lakes (0.50 to 1.00) are evident in the southwest corner of island, again likely owing to glacial dispersal of overburden with buffering potential greater than that demonstrated for most of the region. Jefferies (1986) had found Labrador (owing to low rates of acid deposition) and northeastern Ontario (high CaCO content in overburden) to demonstrate the highest ratios in eastern Canada, while the ratio was consistently lower for lakes in parts of Atlantic Canada than in Quebec and Ontario. Nova Scotia had the lowest median ratio for all of eastern Canada (30% of its lakes were less than 0.00). Nova Scotia, New Brunswick, and insular Newfoundland had a significant proportion of freshwaters with negative ratios reflecting the extremely low availability of CaCO, in the lake catchments in these regions. Jefferies (1986) and Howell and Brooksbank (1987) have cautioned that the Alk: (Ex.Ca 2 + Ex.Mg $^{+}$ 2) ratio is unable to distinguish between natural (organic) and anthropogenic effects.

Excess Sulphate: Excess Calcium and Magnesium

This ratio incorporates parameters that are indicative of both regional geochemistry and magnitude of acidic deposition. The Ex.SO₄⁻²:(Ex.Ca⁺² + Ex.Mg⁺²) ratio will ideally approach, but never equal, zero in low depositon areas as there are always background concentrations of SO₄⁻² in freshwaters (Harvey et al. 1981, Jefferies et al. 1986). The ratio will near unity (1.0) as SO₄⁻² replaces alkalinity in higher deposition regions, and may exceed unity in very high deposition areas. The Ex.SO₄⁻²:(Ex.Ca⁺² + Ex.Mg⁺²) ratio is considered to be relatively less affected by the presence of organic acids (COOH) (Jefferies 1986).

The spatial distribution of Ex.SO₄⁻²:Ex.Ca⁺² + Ex.Mg⁺² ratios for the study lakes is presented in Fig. 14. It is evident that many of lakes with higher ratios (0.51 to 1.00 and greater) lie in the western portion of the study area while lower ratios (less than 0.20) are more frequent in the eastern subregion. This is consistent with the west to east gradient in lake sulphate values (Fig. 7) suggesting a slight gradient in sulphate deposition. It follows that lakes in the western subregion are experiencing a higher rate of alkalinity loss, as sulphate replaces bicarbonate, than are lakes further to the east.

(1986) also Jefferies demonstrated southwest to northeast gradient in this ratio for insular Newfoundland and suggested the trend likely represented a deposition gradient. Regionally, lowest median values for this ratio were evident in northern Ontario (due to high base cation concentrations) and in Labrador (low sulphate levels). Highest ratios were evident in sensitive and/or high deposition regions including south-central Ontario, southern Quebec and parts Atlantic Canada. Most of oundland, the Avalon Peninsula, southern Newfoundland. and the eastern portion of the Northern demonstrated higher ratios (0.6 and greater), reflecting the extremely low base cation levels in these areas, as median lake sulphate values for the island were among the lowest in eastern Canada. The proportion of higher ratio lakes, and geographic distribution of values, supports other evidence indicating that insular Newfoundland, despite the geographical distance from sources of acidic pollution, is still being impacted by acidic deposition, largely owing to extreme terrain sensitivity.

Alkalinity: Excess Sulphate

This ratio is considered to provide a direct indication of the acidification process, reflecting the replacement of alkalinity by sulphate. The ratio can be very large in relatively insensitive terrain and low SO_4^{-2} deposition areas (greater than 10.0) and can range to less than zero (0.0) when alkalinity is exhausted. Important ratio values are considered to be 1.0 (Alk. = Ex.SO₄⁻²), 0.5 (Ex.SO₄⁻² = 2 X Alk.), and 0.2 (Ex.SO₄⁻² = 5 X Alk.) (Jefferies 1986).

The spatial distribution of Alk:Ex.SO₄⁻² ratios for the study lakes is presented in (Fig. 10) and has been previously discussed (as the HCO₃/Ex.SO₄⁻² ratio). The broad scale regional trends in this ratio follow the same patterns as those presented for the other two ion ratios (Jefferies 1986). Jefferies (1986) reported Newfoundland to have a median Alk:Ex.SO₄⁻² ratio of 1.1, which is higher than the more heavily impacted regions (Nova Scotia, southern New Brunswick, south-central Ontario and Quebec) but lower than other regions (Labrador, Northern Ontario). High values of this ratio in association with limestone deposits on the Northern Peninsula and in western Newfoundland have resulted in this island-wide median value being higher than expected.

WRIGHT-HENRIKSEN ACIDIFICATION EQUATION

Henriksen (1982) and Wright (1983) have developed the empirical considerations used to derive the 'nomograph' into a mathematical expression of the acidification process, which provides two quantitative measures of the degree of acidification. The empirical model derives from an ionic balance (with sea salt correction) where:

$$H^{+}+A1^{+}$$
, $+Ex.Ca^{+}$, $+Ex.Mg+$, $+Ex.Na^{+}+Ex.K^{+}+NH_{4}^{+}=NO_{3}^{-}$, $+Ex.SO_{4}^{-}$, $+COOH^{-}$ (1)

In dilute, oligotrophic, clear waters minor ions (NH₄⁺, NO₃⁻², COOH⁻) can be neglected. The model defines one measure of acidification as the loss of alkalinity (which is the original alkalinity minus present day alkalinity). The second measure of acidification, defined as "net" sulphate, is the amount of excess sulphate above background. If the assumptions implicit in model development are correct, then the two measures of acidification are equal as expressed in the Wright-Henriksen acidification equation:

The empirical model was tested on 15 lake data bases from North America and 12 data sets from Europe and proved valid for more than 90% of the oligotrophic, soft-water, clear lakes examined by Wright (1983).

The original (Henriksen 1980) empirical model for lake acidification is static, but the empirical relationships describing the titration model do provide a basis for predicting lake acidification trends given changes in deposition (as sulphate) scenarios. The measure of acidification provided by net $\operatorname{Ex.So}_4^{-1}$ can be arbitrarily set to examine any possible scenario in sulphate deposition, be it an increase or decrease, where:

$$\Delta Net \ Ex.SO_4^{-2} = Ex.SO_4^{-2}(p) - Ex.SO_4^{-2}(t)$$
 (3)

p = predicted (or specified), t = today

The Wright-Henriksen acidification equation becomes a titration model such that:

$$\Delta \text{Net Ex.so}_4^{-2} = \Delta (\text{H}^+ + \text{Al}^{+3} - \text{HCO}_3^-) + \Delta$$

$$0.91 (\text{Ex.Ca}^{+2} + \text{Ex.Mg}^{+2})$$
(4)

In examining a scenario of increased or progressing acidification, an important consideration is as to whether lake acidification is accompanied by increased rates of weathering of base cations in addition to alkalinity loss. The ratio of change in base cations (excess calcium and excess magnesium) to change in excess sulphate is defined as:

$$F = \frac{\Delta(Ex.Ca^{+2} + Ex.Mg^{+2})}{\Delta Ex.So_4^{-2}}$$
 (5)

Henriksen (1982) addressed the issue by comparing historical and recent lake chemistry data (that included measurement of calcium, magnesium, and

sulphate) and by evaluating ranges in calcium and magnesium concentrations for lakes with similar surficial/bedrock geology over a gradient in acid loadings. There was no clear answer to whether base cation levels increased in response to acid loading, however, Henriksen did estimate a maximum base cation increase factor (F) of 0.4. The factor (F) can be set anywhere between 0 and 0.4 at the discretion of the individual(s) applying the model. The factor (F) is incorporated into the acidification equation as follows:

$$\Delta \text{Ex.SO}_4^{-1} = (1-\text{F}) \times \Delta (\text{H}^+ + \text{Al}^+ - \text{HCO}_3^-) + \text{F}$$

$$\times \Delta 0.91 (\text{Ex.Ca}^{+1} + \text{Ex.Mg}^{+1}) \qquad (6)$$

Future (predicted) base cation levels can be calculated as follows:

$$(Ex.Ca^{+2}+Ex.Mg^{+2})_{p} = (Ex.Ca^{+2}+Ex.Mg^{+2})_{t} + F \times \Delta Ex.So_{A}^{-2}$$
 (7)

where p = predicted, t = today.

Similarly the predicted total of hydrogen ion $\{H^+\}$, ionic aluminum $(Al+^3)$, and bicarbonate (HCO,) is as follows:

$$(H^{+}+A1+^{3}+COx)_{p}=(H^{+}+A1+^{3}+CO_{3}^{-})_{t}+$$

$$(1-F)\times\Delta Ex.SO_{4}^{-2}$$
(8)

The acidification equation then can be reduced to calculate future bicarbonate (HCO₃) levels for given changes in excess sulphate as follows:

$$HCO_3^-(p) = HCO_3^-(t) + [F \times 0.91]$$

 $\times \Delta(Ex.Ca^{+2} + Ex.Mg^{+2})] - \Delta Ex.SO_4^{-2}$ (9)

Predicted lake pH can be calculated from HCO₃ (p) (after Wright 1983):

$$pH(p) = 11.3 + log (HCO_3)(p)$$

As a result of this predictive titration model, the projected alkalinity (HCO₃) and pH of the south coast study lakes can be determined for projected scenarios of sulphate deposition and for arbitrarily set values of the base cation increase factor (F). The basic assumption is that current loadings approximate a rainfall pH of 4.7 and SO₄ loadings of 20 kg ha⁻¹ yr⁻¹, and that lake sulphate averages 40 µeq L⁻¹. Three scenarios of increased acid loadings are examined as follows:

- (1) $\Delta \text{Ex.SO}^{-2}_{2} = 20 \, \mu \text{eg L}^{-1}; \text{ x lake}$ $50_{4}^{-2} = 60 \, \mu \text{eg L}^{-1}; 50\% \text{ increase over}$ current (assumed) acid loadings
- (2) $\Delta \text{Ex.SO}_{4}^{-1} = 40 \, \mu \text{eg L}^{-1}$; x lake SO₄ 80 $\mu \text{eg L}^{-1}$; 2 times assumed loadings
- (3) $\Delta \text{Ex.SO}_4^{-1} = 100 \ \mu \text{eq L}^{-1}$; x lake SO $_4^{-2}$ $^{-1}40 \ \mu \text{eq L}^{-1}$; loading are 3.5 times assumed loading, approaching those in regions of southern Quebec and Ontario.

The base cation increase factor (F) has been set at F=0.0 and F=0.4. Histograms of frequency distributions of projected (predicted) lake pH values for south coast lakes under the combined

changes in excess sulphate and ${\tt F}$ factor scenarios are presented in Fig. 16.

The potential effect of increases in acidic deposition on the study lakes is readily apparent. Currently, 7% of the study lakes are in the pH range 4.5 to 5.0 (acidified) and a 50% increase in sulphate loading would result in 32% of the lakes becoming acidic (less than pH 5.0) and 2% of the lakes becoming extremely acidic (less than pH 4.5). Greater increases in acid loading would result in a higher proportion of lakes undergoing acidification. A doubling of current sulphate loading (to 80 μ eq L $^{-1}$ SO $_4^{-2}$ in lake water) would result in 64% of the south coast lakes demonstrating acidification (33% in the pH 4.5 to 5.0 range, 31% with pH less than 4.5). If south coast lakes were receiving acid loading rates similar to those recorded in southern Ontario and (rainfall pH~4.2, sulphate deposition 40 kg ha⁻¹yr⁻¹, lake sulphate -140 μ eq L⁻¹), 97% of the study lakes would demonstrate acidification (89% in the pH range less than 4.5). examination of the same scenarios using the base cation increase factor (F) set at 0.4, the maximum suggested by Henriksen (1982), results in similar trends to lake acidification (Fig. 16). This dramatically portrays the extreme susceptibility of south coast lakes to potential anthropogenic acidification and further highlights the potential impact that increased rates of acidic deposition might have on this region.

The use of Wright-Henriksen acidification equation to explore potential acidification given various scenarios of increases in acidic deposition (sulphate) is of interest in exemplifying the extreme sensitivity of this study region. However, this type of evaluation is not its limitations. This model is effectively a steady state model, and consequently no time frame can be put on the acidification process. Secondly, this model considers only the bicarbonate buffering systems in natural waters, and does not consider the other potential buffering systems that can become increasingly important as bicarbonate is exhausted (consumed). Clair (1987) has pointed to the potential buffering role of humics in brownwaters below pH 5.5. Malanchuk and Turner (1987) have suggested that most brown water systems, even in high deposition regions, do not go below pH 4.5 due to the buffering potential of weak acid anions (COOH). In clear water systems, aluminium and to lesser extent manganese and other metals, liberated from lake catchments due to the acid neutralizing processes, may also assume important buffering roles below pH 5.0 (Johnson 1979). In addition, the strong influence of the marine environment on freshwater chemistry in south coast lakes has been clearly demonstrated. The Wright-Henriksen model takes no account of marine aerosol driven cation exchange reactions that may be occurring in dilute coastal watersheds. These type of reactions have been demonstrated to result in a misrepresentation of a watershed ability to supply base cations (and consequently bicarbonate alkalinity) (Thompson 1982) and have also been demonstrated to contribute to acidification (Seip and Tollan 1982). episodic Due to these inherent limitations in applying the model for predictive purposes, as outlined in the preceding discussion, the lake pH distributions resulting from projected sulphate deposition increases (Fig. 16) must be considered to represent a 'worst case' scenario.

COMPARISON OF LAKE STATUS WITH NOVA SCOTIA

A comparison of the sensitivity and status of dilute freshwater lakes on the south coast of Newfoundland with lakes in Nova Scotia is warranted. Lakes in these two regions are similar with respect to many biophysical characteristics including their relatively small size, thin non-calcareous overburden, acid sensitive non-soluble bedrock, and the considerable influence of widespread peatlands within watersheds and the humification of soils contributing to natural acidity (Scruton 1985, Howel and Brooksbank 1987). In addition, both regions are characterized by high precipitation/runoff and considerable influence of marine aerosols on precipitation chemistry (Banfield 1983; Underwood 1986).

There is considerable controversy over the sources and causes of acidification of Nova Scotian lakes, with long range transport, local (point source) emissions, and natural (organic) sources identified as major contributors (Watt et al. 1979; Underwood and Josselyn 1980; Howell 1986; Jefferies et al. 1986; Howell and Brooksbank 1987). Precipitation acidity in mainland Nova Scotia is considered to vary from pH 4.5 to 4.7 while sulphate deposition ranges from 20 to 30 kg ha¹ yr¹. A gradient in precipitation pH and sulphate deposition from southwestern Nova Scotia (highest deposition, lowest rainfall pH) to Cape Breton is apparent (Underwood et al. 1987). On a time scale, it is likely Nova Scotia has been receiving acid deposition over a longer time frame and at greater levels than has insular Newfoundland, although long term precipitation chemistry data does not exist to confirm this. A comparison of lakes in Nova Scotia with south coast lakes therefore may give us some insight as to the potential future effects of acid deposition if loadings were to increase marginally, if these lakes were to be subjected to continued acid deposition over a longer time period, and also with respect to consideration of Lingan as a potential (relatively close) point source of sulphur emissions of concern for the south coast.

Lake pH in Nova Scotia has been reported to range from 4.23 to 7.67 ($\bar{x}=5.36$; Kelso et al. 1986), 4.0 to 8.0 (Howell and Brooksbank 1987), 4.4 to 6.7 (Howell 1986), 4.2 to 7.7, (median = 5.2; Jefferies et al 1986), and 4.4 to 7.7 ($\bar{x} = 5.0 \pm 0.6$, Underwood et al. 1986). Several authors (e.g. Howell and Brooksbank 1987 for eg.) have reported a bimodal pH distribution which is considered representative of a region experiencing rates of acid deposition sufficient to titrate available bicarbonate alkalinity. Howell and Brooksbank (1987) report 15% of lakes in Nova Scotia to have a pH of less than 5.0, while Jefferies et al. (1986) reported Nova Scotia to have the highest proportion (51%) of acidic and low acid neutralizing capacity (ANC) lakes in all of Eastern Canada. All authors agree that lake chemistry data sets and documentation of river acidification (Watt et al. 1983) confirm Nova Scotia waters as among the most affected systems in Eastern North America, in relation to the LRTAP

phenomenon, despite atmospheric deposition being close to target loading levels (20 kg ha $^{-1}$ yr $^{-1}$). Further, Nova Scotia lakes have been reported to have average excess sulphate concentrations of from 46 μ eq L $^{-1}$ for a sample of 130 lakes (Jeffries et al. 1986), to 83.7 μ eq L $^{-1}$ for a data base of 234 lakes (Underwood et al. 1986) to a high of 128 μ eq L $^{-1}$ for 9 Halifax lakes (Underwood et al. 1987). The potential effect of similar lake sulphate sulphate values (as an indicator of acid loading rates) on south coast lakes is readily apparent in Fig. 16.

Underwood et al. (1986) have noted that lake and river systems in Nova Scotia are not reacting the same to acid deposition. While the acidification of 9 salmon rivers in the province has been conclusively documented, deleterious effects of acid deposition on lake systems are not as evident. This difference between acidification of lentic and lotic habitats may be in part a question of rate of response. Several authors (e.g. Oliver and Kelso 1985; Kelly et al. 1982; Schindler 1986) have suggested that lake sediments may be an important source of acid-neutralizing capacity in standing waters, largely unconsidered to date.

Kelso et al. (1986) have developed an estimat of freshwater resources at risk from anthropogenic acidification in eastern Canada. The data from 814 Eastern Canadian lakes (chemical data collected in the Fisheries and Oceans' NIS survey program) was applied to an estimated 700,000 lakes considered to be in a 'risk' area (number of lakes estimated from regional counts and measures information). The authors have assumed the risk area to encompass the land mass east of the Ontario-Manitoba boarder and south of 52° latitude, an area considered to be receiving_acid deposition in the order of 20 kg ha⁻¹ yr⁻¹ of SO_A^{-2} or greater. Virtually the whole of Labrador SO₄ or greater. Virtually the whole or Labrador is north of 52° latitude and is classified as a non-risk area. Lake inventory data applied to counts and measures information suggested a total freshwater area of 10,930 kms and a total number of lakes of 138,711 (greater than 1.0 ha in size) the 'risk area' for the province of Newfoundland and Labrador, associated (primarly) with the island of Newfoundland. Kelso et al. (1986) have evaluated the estimated lake distributions in relation to acidification alkalinity values, and pH. For all of insular Newfoundland, it was estimated that 124,701 lakes (89.9%) would be expected to have CSI value of less than -3.00, 75,181 lakes (54.2%) would have extremely low alkalinity (less than 50 μ eq L $^{-1}$), and 41,475 lakes (29.9%) would have a pH of less than 6.0. Some 2,497 lakes (1.8%) would be considered 'acidified' (pH less than 4.7) after Henriksen (1980). It is likely the majority of these 'acidified' lakes are naturally dystrophic.

CONCLUSIONS

The south coast of insular Newfoundland is of concern to Fisheries and Oceans with respect to the potential for acidification damage and adverse effects on freshwater fish. The low buffering potential afforded freshwaters in the region from underlying bedrock and overburden, high annual precipitation inputs, proximity to regional and

long range sources of acid pecursers, and the widespread occurrence of humic deposits, have targeted the south coast as having considerable potential to be impacted from acid deposition. Additionally, the morphometric and hydrological characteristics of the lakes within this region further contribute to their susceptability.

This survey has confirmed the expected lake sensitivity. Despite the fact the survey was conducted during a period when aquatic sensitivity indicators would be expected to demonstrate a 'best case' scenario, the extreme susceptibility of the study lakes is evident. All study lakes alkalinity values below the standard for high sensitivity while 42 lakes (47%) were classified as extremely sensitive (less than 40 μeq L $^{-1}$) and 14 lakes as acidified (no alkalinity). Other chemical indicators (calcium, conductivity, calcite saturation index, etc.) evidence of aquatic demonstrate similar sensitivity. Depletion of buffering capacity is evident in most of the study lakes. Alkalinity deficits ranged from 0.8 to 119.7 μ eq L¹ (x = 33.4) and in 68% of the lakes residual alkalinity is less than the alkalinity loss. Non-marine sulphate values in the study lakes were low in comparison to other parts of eastern Canada, however, sulphate is dominant to (replacing) bicarbonate in 50% of the lakes. Lake values ranged from 4.61 to 6.67 (x = 5.76) while clearwater lakes had values of greater than 5.30. Lake pH distribution was even across the pH range 5.00 to 6.50, suggesting residual alkalinity in most of the study lakes, however, six lakes had pH values of less than 5.00.

The importance of organic contributions to lake chemistry is clearly evident both qualitatively (water colour) and quantitatively (DOC, COOH values). Organic anions (COOH) are the dominant non-marine anionic constituent in 45 lakes (50%). The organic content of the region's freshwaters is contributing to low pH/alkalinity and dystrophy has been previously identified as a key factor controlling the physical and chemical characteristics of insular Newfoundland lakes (Scruton 1985, Earle et al. 1987). The influence of the marine environment on freshwater chemistry is also clearly evident as chloride is the dominant anion in 68 lakes (76%), prior to extracting marine contributions.

Empirical acidification models developed by Henriksen (1979, 1980) suggest ongoing lake acidification, however the models do not distinguish between anthropogenic and natural contributions to the process. Lakes demonstrating acidification on Henriksen's 'Indicator Diagram' (17 lakes, 20%) were coloured to highly coloured, while 3 lakes were sulphate dominated. Henriksen's 'Nomograph' indicated 6 lakes were in 'Transition' to becoming acidified while the remaining lakes fell in the 'Bicarbonate' region of the model. The nomograph incorrectly predicted the pH of 16 (19%) of the study lakes. Due to the preponderance of organic acidity in the study lakes and other assumptions implicit in model development, these two models are considered poor in their applicability to Newfoundland lakes.

Ion ratios, employed by Jefferies (1986) to demonstrate broadscale regional response to acidic deposition, confirmed the high sensitivity of the

south coast region and provided evidence of ongoing lake acidification. The distribution of ion ratio values for south coast lakes points to the critical nature of aquatic sensitivity of freshwaters in the region and the confirmed the process of alkalinity depletion. The ratios point to southern Newfoundland as being an 'acid-impacted' region, however less so than southern Ontario and Quebec, and southwestern Nova Scotia. Organic acidity, while not incorporated into the ion ratios, is not considered to explain the distribution of values on a regional basis (Jefferies 1986).

The Wright-Henriksen acidification model was used to examine scenarios of increased rates of acid loading and the resulting effects on lake pH distributions. A 50% increase in (assumed rate of) sulphate deposition would result in a 25% increase in acidified south coast lakes (exhaustion of HCO-, alkalinity), while a doubling of sulphate deposition would result in 64% of the lakes having a pH of less than 5.0. Rates of acid loading currently experienced in south-central Ontario would have drastic effects on the region's freshwaters.

In conclusion, the extreme susceptibility of freshwaters on the south coast of Newfoundland to acidic deposition has been well established and progressing acidification (loss of buffering capacity) is apparent. A variety of circumstances suggest that this region of the island is where serious damage from acid precipitation, if it is to occur, may become first apparent. Consequently, agencies currently involved in aquatic acid precipitation research and monitoring should pay attention to the south coast of insular newfoundland to periodically evaluate spatial and temporal trends in chemical and biological parameters indicative of acidification.

ACKNOWLEDGMENTS

Thanks are extended to Mr. Peter Downton and Mr. Marvin A. Barnes who participated in the field collection of water samples and analyses of samples at a mobile field laboratory. Water analyses were completed by a contracted analytical laboratory (Ocean Chem. Ltd., Halifax, N.S.). Mr. Reg Tucker supervised data entry onto computer tape and Mr. Don Stansbury completed the programming and data analysis. Mrs. Kal Lynch and Mrs. Janice Lannon typed the manuscript Mr. Herb Mullett prepared the graphics. Mr. Pat M. Ryan and Mr. Marvin A. Barnes constructively reviewed the manuscript.

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Table 1. Water quality parameters measured in the laboratory including analytical methods, equipment used, and detection limits.

Parameter	Analytical procedure/equipment	Detection limi	
рН	Accumet 144 digital pH meter	0.01 pH unit	
Total alkalinity	Two end point determination using Accumet pH meter and Metrohm auto-buret system	0.1 µeq L ⁻¹	
Conductivity	Chemtrix conductivity meter with KCl standards	1.0 μScm ⁻¹	
Total hardness	By calculation from Ca and Mg determinations, expressed as $\mu eq~L^{-1}~CaCO_3$	0.1 $\mu eq L^{-1}$	
DOC (dissolved organic carbon)	Carbon analyzer with infra-red detector	0.1 mg L ⁻¹	
TDS (total dissolved solids)	Weight after removal of water by heating	0.1 mg L ⁻¹	
Colour	Helge colour tester system	5 TCU	
Calcium, magnesium, potassium, sodium, aluminum	Inductively-coupled plasma spectrometer	$0.1 \mu eq L^{-1}$	
Bicarbonate	By calculation from field pH and alkalinity data		
Chloride, sulphate, nitrate	Ion-chromatography	0.1 μ eq L^{-1}	
Sulphate	MTB (reported separately)	0.1 μ eq L^{-1}	
Ortho-phosphate, ammonium	Technicon Auto-analyzer	$0.001 \ \mu eq \ L^{-1}$	
Turbidity turbidimeter	Turner Design, digital 0.1 JTU		

Table 2. Statistical summary of morphometric characteristics and physical/chemical properties for the 90 south coast study lakes.

Parameter		Minimum	Maximum	Mean	Standard deviation	
Morphometric characteristics						
Lake area (ha)	90	3.0	773.5	62.0	122.8	
Watershed area (ha)	90	15.4	5160.0	421.4	696.7	
Watershed area to lake area ratio	90	1.0	87.7	13.7	15.3	
Elevation (m)	90	30.0	495.3	300.8	104.0	
Distance from the coast (km)	90	2.5	80.0	29.1	17.5	
Physical properties						
Colour (TCU)	89	3.5	120.0	47.7	28.5	
Turbidity (JTU)	89	0.06	4.50	0.77	0.48	
Hardness	89	9.9	115.2	38.3	18.0	
Conductivity (µS cm ⁻¹)	89	7.2	76.0	14.0	9.4	
Total dissolved solids (mg L ⁻¹)	89	10.0	305.0	54.0	51.5	
Lake chemistry						
pH (pH units)	85	4.61	6.67	5.76	0.56	
Alkalinity	85	-29.6	154.8	23.7	27.8	
Calcium	89	6.8	158.8	40.5	26.0	
Magnesium	89	10.3	60.3	23.3	10.1	
Sodium	89	30.7	113.1	65.1	13.3	
Potassium	89	0.7	7.9	3.7	1.2	
Hydrogen ion (H ⁺)	85	0.2	24.5	3.6	4.6	
Aluminum (µg L ⁻¹)	85	33.0	554.0	356.7	105.5	
Bicarbonate	90	0.0	188.5	31.3	33.2	
Chloride	89	12.5	62.5	31.5	18.3	
Sulphate	89	12.5	62.5	31.5	11.2	
Orthophosphate	90	0.000	0.184	0.009	0.028	
Anion sum	89	63.3	271.4	138.5	42.3	
Cation sum	89	48.7	280.5	132.5	38.2	
Excess calcium	89	5.7	157.1	38.9	30.0	
Excess magnesium	89	6.9	56.5	18.3	10.2	
Excess sodium	89	0.0	47.1	23.2	9.0	
Excess potassium	89	0.0	6.5	2.2	1.1	
Excess sulphate	89	5.4	44.4	21.0	9.2	

⁻ all parameters in $\mu eq~L^{-1}$ except where specified

Table 3. Order of cationic and anionic dominance (for both total and non-marine concentrations) for south coast lakes, including a breakdown by geological type.

					Geological Code			
		n	<u> </u>	04	06	07	09	
Cations								
Na < Ca < Mg < K		70	(78.7)	23	35	4	8	
Na < Mg < Ca < K			(11.2)	2	5			
Ca < Na < Mg < K			(7.9)		2	1	3 4	
Ca < Mg < Na < K			(2.2)	- 1		_		
J		89	•	$\overline{26}$	42	5	$\frac{1}{16}$	
<u> Von-Mari</u> ne (Excess) Cations							
Ca < Na < Mg < K		43	(48.3)	10	22	5	6	
Ca < Mg < Na < K			(20.2)	3	7	0	8	
Ca < Mg < K < Na			(1.1)	1	_	-	-	
Na < Ca < Mg < K			(24.7)	12	10	_	_	
Mg < Ca < Na < K			(3.4)	-	2	-	1	
Mg < Na < Ca < K			(2.2)	-	1	<u>-</u> 5	$\frac{1}{16}$	
		89		26	42	5	16	
Aniona								
Anions	COOR	10	(12.2)	2	2		1	
$C1 < BCO_3 < BCO_4 <$	COOR		(12.3)	3 2	2 3	4 1	1	
C1 < HCO ₃ < SO ₄ < C1 < SO ₄ < HCO ₃ < C1 < SO ₄ < C00H < C1 < C00H < C1 < C00H < C1 < C00H < C1 < C1 < C00H < C1 < C	HCU COOU		(7.4) (21.0)	8	3 8	1	1	
C1 < COUH < CO <	HCO3		(30.9)	8	14	_		
C1 < COOH < SO, <	503		(6.2)	1	4	_	3	
C1 < HCO3 < COOH <	504		(6.2)	1	4	_	_	
соон < c1 < so ₄ <	HCO.		(2.5)	_	1		1	
COOH < C1 < HCO. <	S0.		(6.2)		1	_	4	
соон < с1 < нсб ₃ < соон < нсо ₃ < с1 <	S0,		(1.2)	_	1	_	_	
$HCO_{3} < C1 \stackrel{?}{<} SO_{4} <$	COOH		(1.2)	_	_	_	1	
HCO3 < Cl < COÖH <	SO,		(3.7)	1	1	_	1	
HCO ₃ < C1 < SO ₄ < HCO ₃ < C1 < COOH < HCO ₃ < COOH < C1 <	: so ⁴		(1.2)	-	1	-	_	
3	4	81	•	24	40	5	12	
Non-marine (Excess) Anions	2	(2.7)	1		0		
$\frac{100_{3} < 50_{4} < 600H}{100_{3} < 600H} < 50_{4}$			(3.7)	1	10	2	_	
SO_4 < COOH < HCO ₃			(23.5)	4 3	10	2	3	
$SO_4 < HCO_3 < COOH$			(12.3) (4.9)	2	6 1	0 1	1	
$cooh < so_4^3 < hco_3$			(33.3)	11	13	_	3	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$			(22.2)	3	10	_	5	
3 . 554		$\frac{30}{81}$	(22,2)	24	40	5	$\frac{3}{12}$	
Key								
Geological code	Bedrock type							
04		, a	nd foliated granite.				•	
06	granite.							
07			pyrienite, granodiori	te				
00	and mafic syeni							
09			canic rocks with asso	clated				
	minor rock type	=5						

Table 4. A comparison of marine contributions to freshwater chemistry from this study with an island-wide survey of lakes (Scruton 1983), Labrador lakes (Scruton 1984) and insular Newfoundland rivers (Scruton 1986). All values in $\mu eg~L^{-1}$.

	South Coast lakes (n = 90)		Insular Newfoundland lakes (n = 109)		Labrador lakes (n = 130)		Insular Newfoundland rivers (n = 22)	
	Value	%	Value	%	Value	%	Value	%
Chloride	75.4	100	87.8	100	9.4	100	116.0	100
Sodium	65.1	64	82.3	60	22.1	35	100.9	64
Potassium	3.7	41	3.3	55	3.2	5	6.3	44
Sulphate	31.5	33	46.8	26	28.2	3	59.3	27
Magnesium	23.3	21	50.2	12	32.2	6	40.4	19
Calcium	40.5	4	102.5	<1	60.6	<1	64.5	4

Table 5. Sensitivity of study lakes to acidification based on various criteria.

		Sensitivity to acidification	n	<u>%</u>
1.	Alkalinity ($\mu eq L^{-1}$) ^a			
	less than 0 0-40 40-200 200-500	acidified extremely sensitive highly sensitive	14 42 19	16.5 49.4 22.4
	500+	moderately sensitive not sensitive	-	_
2.	Calcium $(\mu eq L^{-1})^b$ $0-200$ $200-400$ greater than 400	highly sensitive moderately sensitive least sensitive	89 - -	100.0
3.	Conductivity (µScm ⁻¹) ^c			
	0-35 22-78 greater than 60	highly sensitive moderately sensitive least sensitive	85 12 1	95.5 13.5 1.1
4.	Calcite Saturation Index (CSI) ^d			
	less than -3 -2 to -3 -1 to -2 greater than -1	deleterious effects possible danger moderate sensitivity low sensitivity	70 - - -	100.0
5.	Bicarbonate to Excess Sulphate I	<u>Ratio</u> e		
	$\geq 1.0 \text{ (HCO}_{3^{2}} \geq \text{SO}_{4}^{-2})$ $< 1.0 \text{ (SO}_{4}^{3^{2}} \geq \text{HCO}_{3}^{-1})$	moderate to low sensitivity buffering ability depleted	42 42	
6.	Non-marine Calcium and Magnesium	n vs. Bicarbonate ^b		
	at equivalence line below equivalence line below equivalence line with	<pre>non-impacted impacted or 'acidifying'</pre>	3 81	3.6 96.4
	negative alkalinity	acidified	14	16.7
7.	pH-Calcium Relationship ^f above empirical curve below empirical curve	acidified non-acidified	20 64	23.8 76.2
			. Co	nt'd.

Table 5 (Cont'd.)

		Sensitivity to acidification	n	%
8.	Henriksen's Nomograph			
	bicarbonate zone	intact buffer system	83	93.3
	transition zone	depletion of buffering capacity	6	6.7
	acidified zone	loss of buffer system	0	0
	Alkalinity to Excess Calci	um and Magnesium ^h		
	less than O	acidified (no alkalinity)	11	12.9
	0.00-0.20	greater than 80% loss in alkalinity	17	20.0
	0.21-0.50	50 to 80% loss in alkalinity	32	37.6
	0.51-1.00	0 to 50% loss in alkalinity	25	29.4
0.	Ratio of Excess Sulphate t	o Excess Calcium and Magnesium		
	greater than 1.00	sulphate is replacing alkalinity	6	6.7
	0.51 to 1.00	alkalinity is up to 2X sulphate	32	36.0
	0.21 to 0.50	alkalinity is from 2 to 5X sulphate	31	34.8
	less than 0.20	alkalinity is greater than 5X	20	22.5
		sulphate		
1.	Ratio of Alkalinity to Exc	ess Sulphate ^h		
	less than 0.0	acidified	12	14.3
	0.00 to 0.20	sulphate is greater than	6	7.1
		5X alkalinity		
	0.21 to 0.50	sulphate is from 2 to	12	14.3
		5X alkalinity		
	0.51 to 1.00	sulphate ranges from equal to	15	17.9
		up to 2X alkalinity		
	1.01 to 10.00	alkalinity is up to 10% sulphate	38	45.2
	greater than 10.00	alkalinity is greater than 10X sulphate	1	1.2

aOntario Ministry of the Environment (1981)
bHarvey et al. (1981)
cGorham (1978)
dKerekes and Hartwell (1980)
eOden (1976)
fHenriksen (1979)
gHenriksen (1980)
hJefferies (1986)

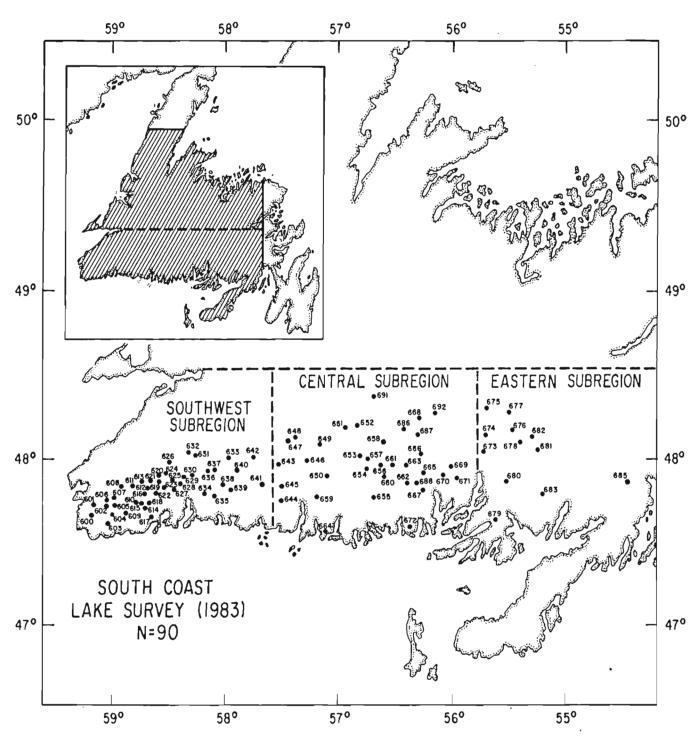


Fig. 1 Map of the south coast of insular Newfoundland showing the location of the study lakes and the geographical sub-regions delineated for this study.

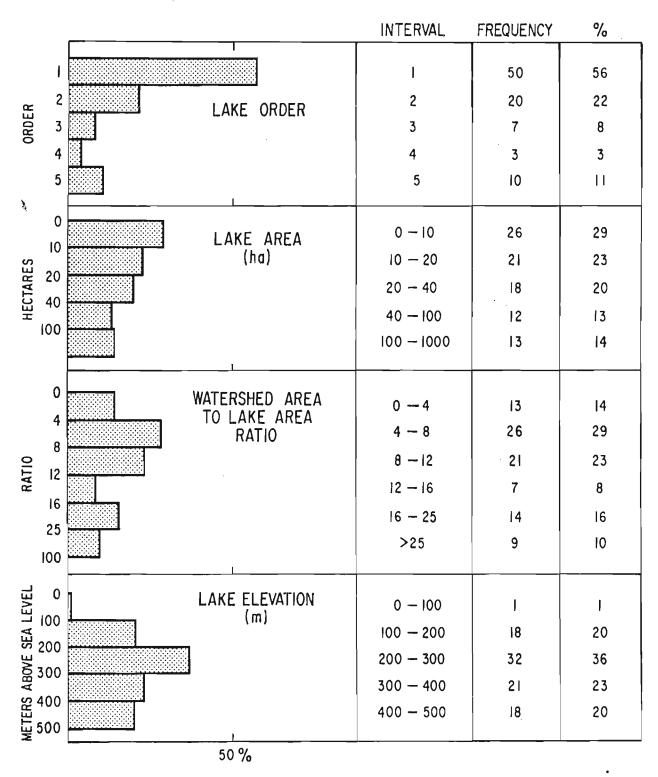


Fig. 2. Histograms of the distribution of key morphometric variables including drainage order, lake area (ha), watershed area to lake area ratio, and elevation (m above sea level) for the 90 study lakes.

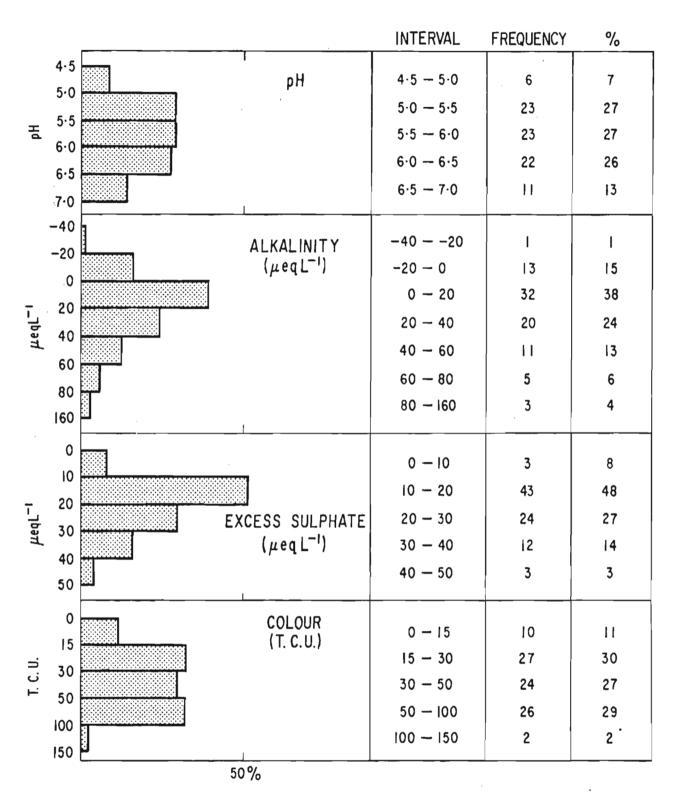
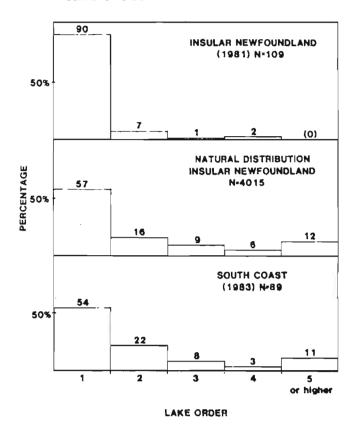


Fig. 3. Histograms of the distribution of key chemical variables including pH, alkalinity ($\mu eq~L^{-1}$), excess sulphate ($\mu eq~L^{-1}$) and water colour (TCU) for the 90 study lakes.

DISTRIBUTION OF LAKES BY ORDER FOR INVENTORY SURVEYS AS COMPARED TO NATURAL DISTRIBUTIONS



DISTRIBUTION OF LAKE AREAS FOR INVENTORY STUDIES AS COMPARED TO NATURAL DISTRIBUTIONS

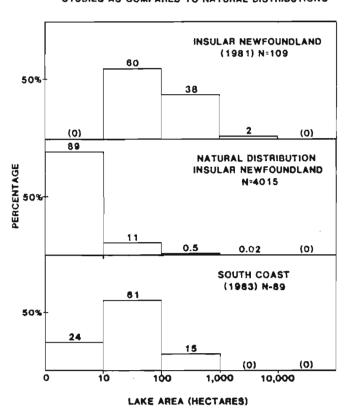


Fig. 4. Study lake distribution by drainage order and by size class (ha) in comparison to natural lake distributions.

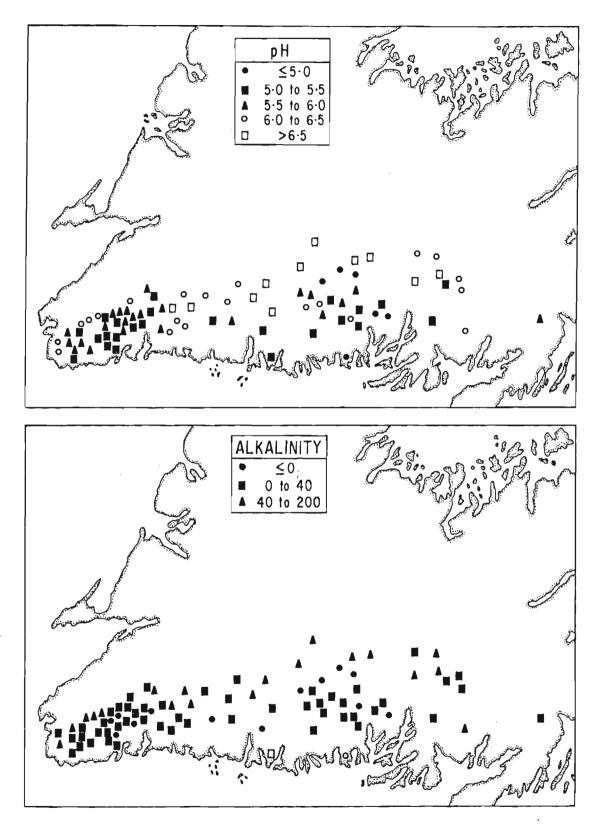


Fig. 5. The spatial distribution of pH and alkalinity ($\mu eq\ L^{-1}$) in the study lakes.

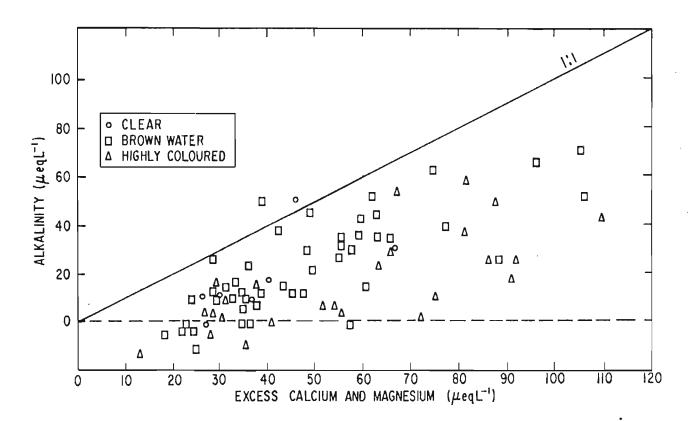


Fig. 6. A plot of excess calcium and magnesium versus alkalinity as an indicator of alkalinity depletion (deficit).

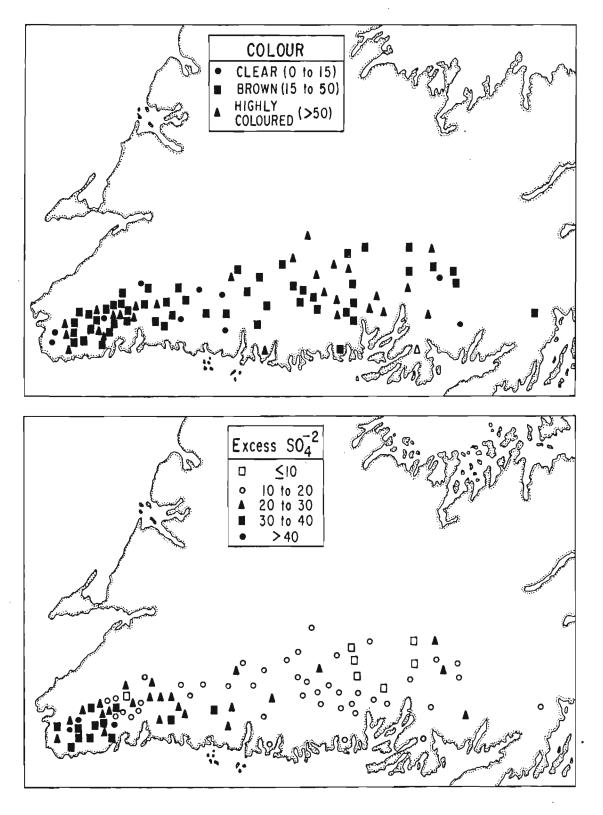


Fig. 7. The spatial distribution of water color (TCU) and excess sulphate ($\mu eq~L^{-1})$ in the study lakes.

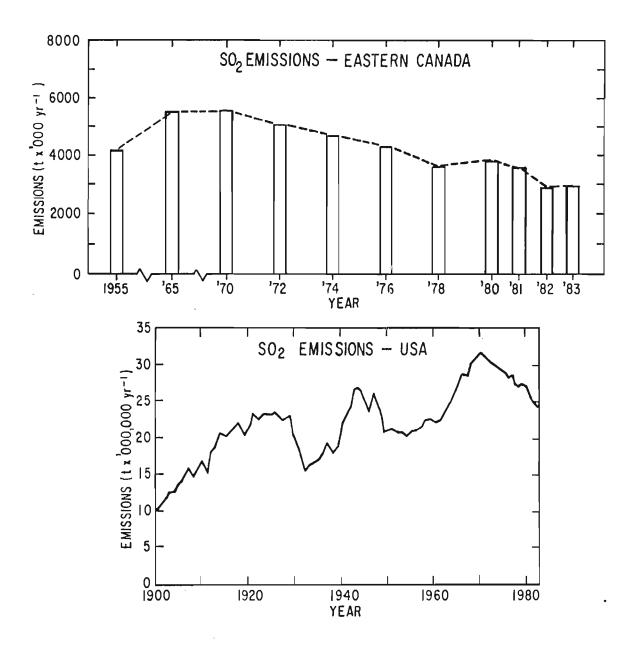


Fig. 8. Temporal patterns in sulphur dioxide (${\rm SO}_2$) emissions from Eastern Canada and the United States (adapted from Martin and Brydges 1986).

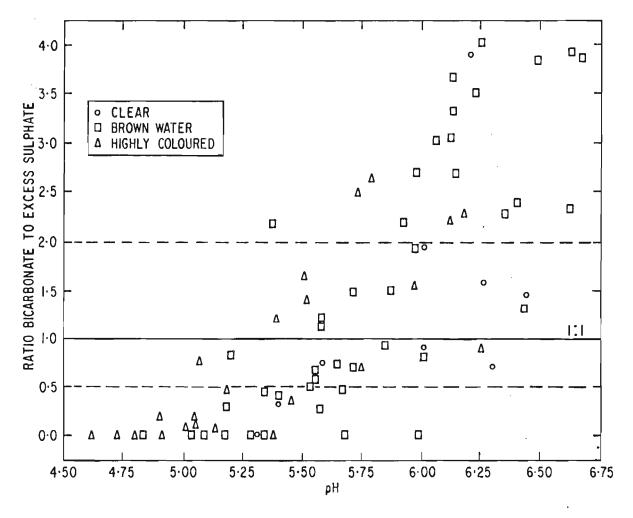


Fig. 9. A plot of the ratio of bicarbonate to excess sulphate (HCO $_3$ -:Ex.SO $_4$ -2) versus pH in the study lakes as an indicator of sulphate replacing bicarbonate.

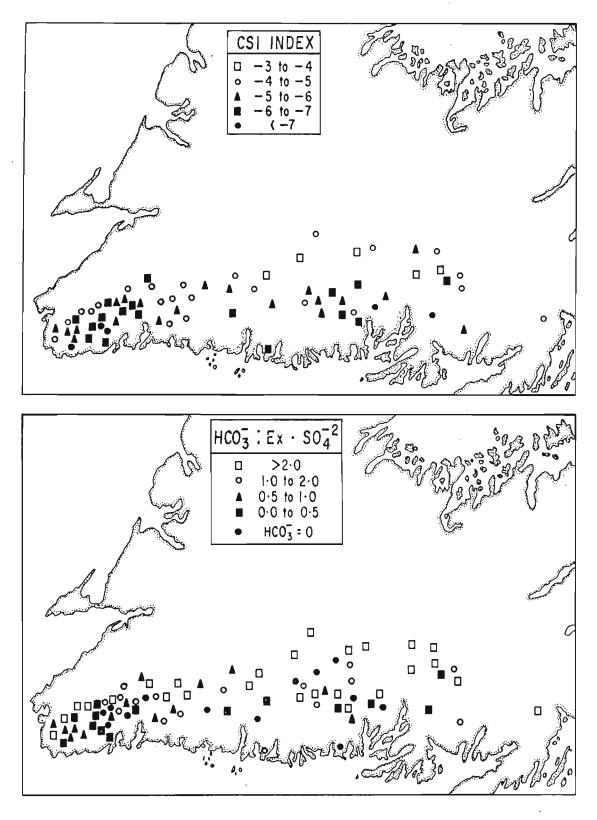


Fig. 10. The spatial distribution of the values of calcite saturation index (CSI) and the ratio of bicarbonate to excess sulphate in the study lakes.

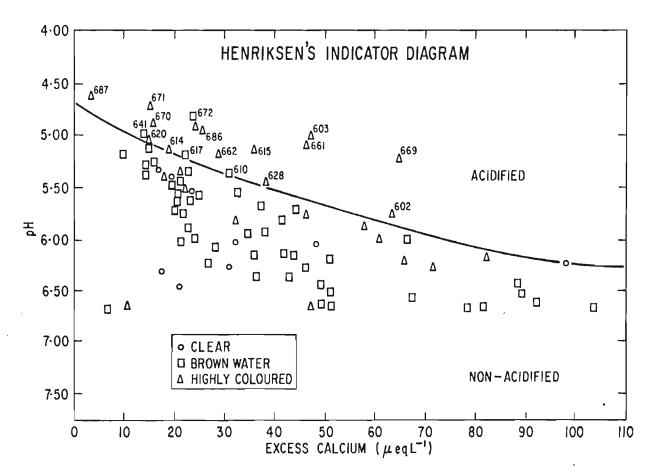


Fig. 11. A plot of excess calcium ($\mu eq~L^{-1}$) and pH data from the 90 study lakes on Henriksen's (1979) 'Indicator Diagram.'

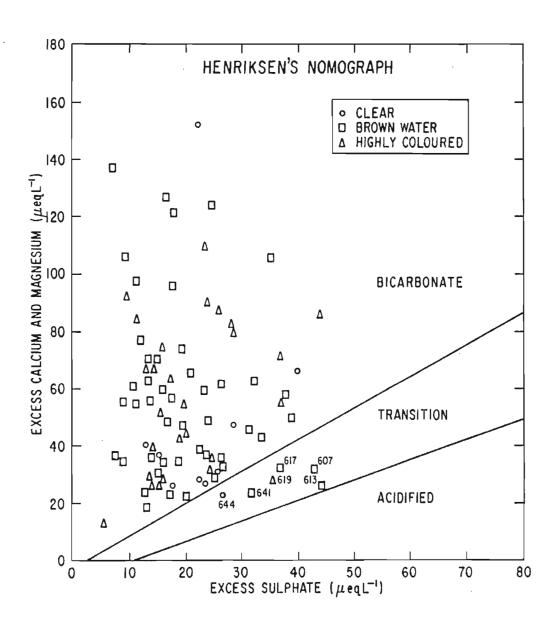


Fig. 12. A plot of excess calcium, magnesium, and sulphate ($\mu eq~L^{-1}$) for the 90 study lakes on Henriksen's (1980) 'Nomograph'.

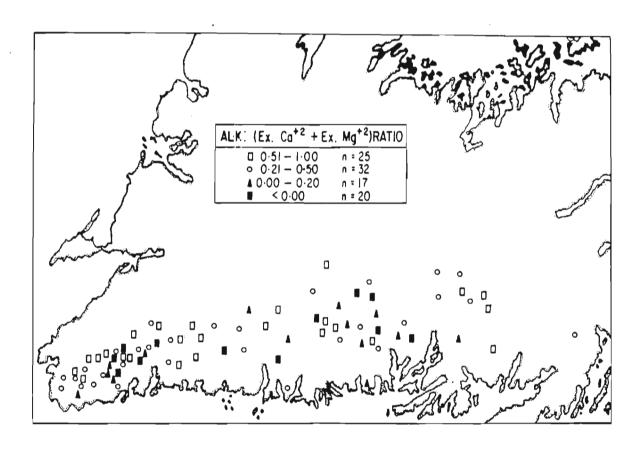


Fig. 13. The spatial distribution of the ratio of alkalinity to the base cations, calcium and magnesium (Alk: Ex. Ca $^{+2}$ + Ex. Mg $^{+2}$), for the 90 study lakes.

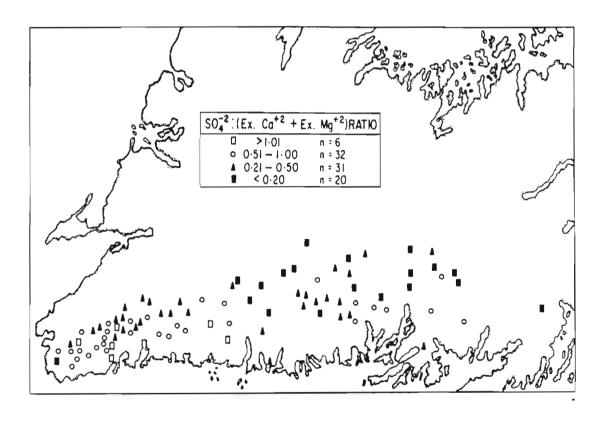


Fig. 14. The spatial distribution of the ratio of sulphate to the base cations, calcium and magnesium (Ex.SO $_4^{-2}$: Ex.Ca $^{+2}$ + Ex.Mg $^{+2}$), for the 90 study lakes.

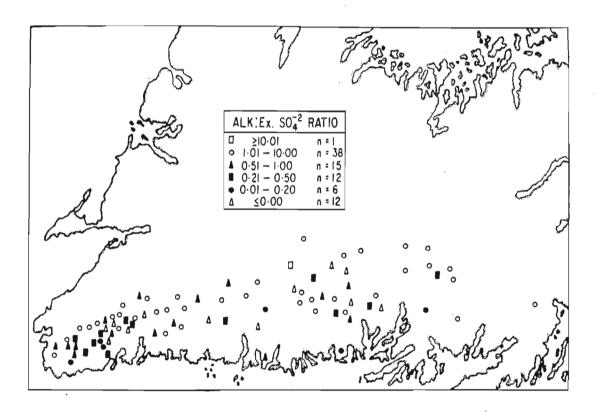


Fig. 15. The spatial distribution of the ratio of alkalinity to excess sulphate (Alk: ${\rm Ex.S0_4}^{-2}$) for the 90 study lakes.

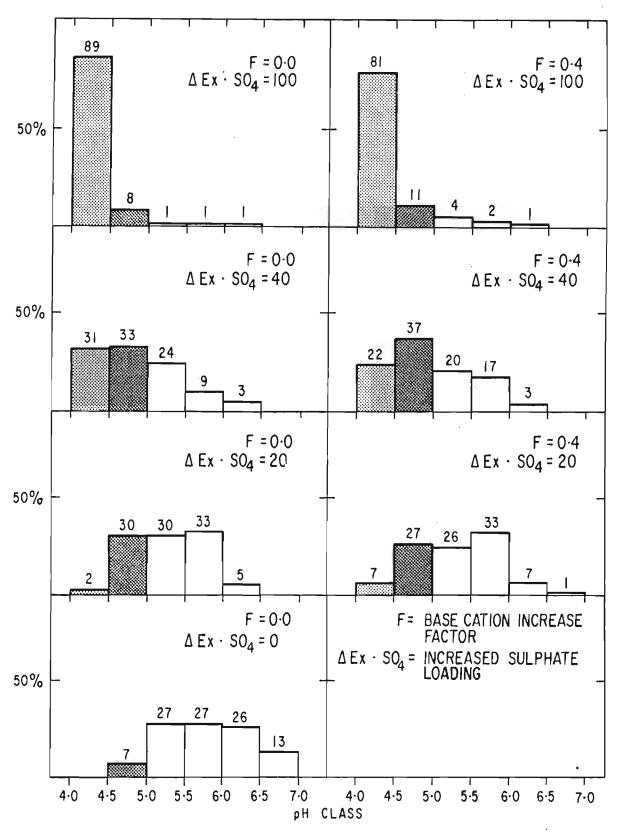


Fig. 16. Projected pH distributions of south coast lakes for given scenarios of increases in acidic deposition (increased sulphate loading or $\Delta \text{Ex.SO}_4^{-2}$) and for two possibilities of increased rates of weathering (base cation increase factor or F). The lower left hand panel presents the current lake pH distribution.

Appendix 1

Morphometric, Physical and

Chemical Data for the Study Lakes

Table 1.1 Siting information and morphometric data for the study lakes.

Table 1.2 Physical and chemical data for the study lakes.

Table 1.1 Siting information and morphometric data for the study lakes (see end of table for explanation of headings and units).

1

SOUTH COAST LAKES - MORPHOMETRIC DATA

LAKE RIVER LAT LONG DATE ORDER GEOLOGY LAREA DAREA RATIO ELEV DISTCST 3920100 4742 5908 830724 3 4 83.4 1266.9 15.2 30.0 9.5 601 3920100 4744 5906 830724 1 15.0 245.1 16.3 142.5 14.0 3920100 4744 5901 830724 50.0 7.5 602 1 9 6.7 160.0 14.8 603 3919820 4738 5902 830724 5 4 11.1 238.4 21.5 129.5 7.0 3919820 4740 5859 830724 2 15.0 246.7 16.4 6 213.4 10.3 605 3919820 4743 5858 830724 1 4 6.0 85.0 14.2 288.8 15.0 606 3919820 4744 5902 830724 1 4 20.0 146.0 7.3 304.8 18.0 607 3919820 4745 5858 830724 1 4 6.0 60.0 10.0 358.1 17.5 608 3919820 4749 5855 830724 4 7 101.0 781.8 77.4 365.8 24.5 3919690 4741 5854 830724 1 9.5 85.0 8.9 236.2 609 6 10.0 3919590 4744 5849 830724 411.5 610 1 11.7 173.4 14.8 14.8 4 3919590 4749 5851 830724 2 7 358.1 9.3 78.3 8.4 23.5 611 1 40.0 153.4 464.8 3919596 4748 5841 830724 6 20.5 612 3.8 3919590 4751 5844 830724 5 388.6 28.3 2482.2 87.7 28.5 613 6 3919460 4743 5842 830724 2 246.7 26.7 312.4 9.5 614 4 9.2 3919460 4744 5845 830724 2 75.0 25.0 297.2 615 4 3.0 14.8 3919460 4747 5842 830724 1 33.3 449.6 616 6 183.4 5.5 18.5 617 3920110 4741 5836 830724 1 6 11.0 90.0 8.2 205.7 2.5 618 3920110 4743 5841 830724 5 76.7 443.2 297.2 5.8 80.0 3819120 4752 5834 830724 1 495.3 619 6 5.0 60.0 12.0 26.5 3819120 4753 5834 830724 620 3 24.0 286.7 11.9 495.3 29.0 6 3819120 4753 5837 830724 2 53.3 376.7 7.1 449.6 621 6 28.0 3819100 4748 5836 830724 622 1 6 16.7 125.0 7.5 388.6 18.0 623 3819100 4750 5832 830724 45.0 311.2 6.9 419.1 12.3 6 3819100 4754 5832 830724 36.7 521.8 14.2 495.3 624 6 20.5 3819100 4752 5828 830724 10.0 100.0 10.0 373.4 625 1 6 25.0 3819100 4757 5829 830724 626 1 7 46.7 436.7 9.4 495.3 27.0 297.2 627 3819030 4748 5827 830724 1 6 41.7 305.1 7.3 11.5 3818860 4751 5823 830724 628 6 14.0 1120.0 80.0 312.4 14.5 629 3818860 4751 5823 830724 1 6 6.0 62.0 10.3 312.4 18.0 630 3818800 4754 5813 830724 1 6 32.0 246.0 7.7 434.3 18.5 631 3818800 4801 5812 830724 4 7 12.0 426.0 35.5 449.5 32.0 3818800 4804 5816 830724 1 4 190.0 630.0 3.3 480.1 34.5 632 3818800 4800 5758 830724 2 4 6.0 120.0 20.0 464.8 38.5 633 9 3818760 4747 5810 830724 1 10.0 248.0 24.8 266.7 10.0 634 175.3 3818490 4746 5803 830724 1 6 28.0 188.0 6.7 6.5 635 134.0 16.8 358.1 26.5 636 3818490 4756 5808 830724 6 8.0 3818490 4756 5801 830724 1 16.0 98.0 6.1 495.3 29.0 637 6 3818340 4752 5758 830724 60.0 190.0 434.3 19.0 638 1 4 3.2 639 3818190 4749 5754 830724 1 6 7.0 116.0 16.6 358.1 14.0 403.9 28.0 640 3818190 4756 5752 830724 1 150.0 5.4 27.5 3817890 4750 5738 830720 100.0 240.0 2.4 358.1 24.0 641 1 6 3817890 4800 5744 830724 110.0 406.0 449.6 642 1 3.7 39.5 6 3817890 4757 5731 830720 2 604.0 2704.0 4.5 358.1 34.5 643 3717770 4744 5731 830720 2 4 228.0 251.5 . 10.0 644 26.0 8.8

Table 1.1 (Cont'd)

SOUTH COAST LAKES - MORPHOMETRIC DATA

LAKE	RIVER	LAT	LONG	DATE	ORDER	GEOLOGY	LAREA	DAREA	RATIO	ELEV	DISTCST
645	3717440	4749	5727	830720	1	4	10.0	118.0	11.8	342.9	20.5
646	3717440	4758	5716	830720	1	4	8.0	146.0	18.3	327.7	37.5
647	3717440	4804	5725	830720	1	9	18.0	196.0	10.9	297.2	39.0
648	3717440	4805	5722	830720	5	6	514.0	1033.0	20.1	266.7	36.0
649	3717440	4802	5708	830720	1	6	26.0	332.0	12.8	373.4	49.0
650	3716620	4753	5704	830720	3	9	11.1	400.0	36.0	297.2	31.0
651	3716220	4808	5654	830719	1	6	8.0	36.0	45.0	327.7	63.0
652	3716220	4809	5648	830722	2	6	32.0	692.0	21.6	251.5	64.0
653	3716220	4800	5647	830719	5	6	280.0	1202.0	4.3	281.9	46.0
654	3716220	4755	5644	830719	2	6	8.0	282.0	35.3	251.5	37.0
655	3615270				5	6	214.0	834.0	3.9	312.4	20.0
656	3615270				1	6	11.0	76.0	6.9	281.9	41.0
657	3615270				1	6	22.0	168.0	7.6	281.9	48.0
658	3615270				1	4	6.0	160.0		297.2	61.0
659	3716960				5	4	12.0	114.0		281.9	21.0
660	3615220				2	6	164.0	752.0	4.6	266.7	27.5
661	3615220				2	4	26.0	312.0		281.9	38.0
662	3614850				1	6	18.0	102.0	5.7	190.5	23.0
663	3614850				3	6	24.0	524.0		297.2	38.0
664	3716860				1	4	65.0	218.0	3.4	236.2	3.5
665	3614260				2	6	26.0	308.0		205.7	32.0
666	3614260				4	4	236.0	848.0	3.6	251.5	42.0
667	3614260				2	4	12.0	172.0		251.5	17.0
668	3614070				1	9	104.0	604.0		251.5	64.0
669	3614070				1	6	238.0	554.0		175.3	18.0
670	3614070				1	4	16.0	144.0		236.2	12.0
671	3614070				2	6	6.0	15.4	2.6	175.3	28.0
672	3615040				2	4	10.0	110.0		312.4	8.0
673	3613410				1	9	6.0	100.0		175.3	47.0
674	3613350				5	9		2248.0		190.5	57.0
675	3613350				3	9	88.0	376.0	4.3	251.5	73.0
676	3613350				3	9	16.0	520.0		190.5	57.0
677	3613350				1	9	4.0		13.0	190.5	59.0
678	3613110				1	9	26.0	108.0	4.2	190.5	49.0
680	3410790 3410790				2	6	4.0		20.0	213.4	9.0
681					1	4	30.0	242.0	8.1	175.3	41.0
682	3410790 3409780				1		60.0	66.0	9.7 5.5	190.5 137.2	49.0 14.0
683 685	3004480				1 1	4 9	16.0	85.0	5.3	175.3	18.0
686	3614260				1	6	10.0	100.0	1.0	312.4	62.5
687	3614200				1	6	5.0	50.0	1.0	274.3	52.0
688	3614070				1	6	5.0	50.0	1.0	274.3	25.0
691	3614070				5	6		5160.0	6.7	182.8	55.0
692	3614070				5	9	140.0	584.0	4.2	480.1	26.0
074	20140/0	4014	2000	000/13	J	,	140.0	204.0	7.4	-00.I	20.0

)

Table 1.1 (Cont'd)

Key to parameter names and units of expression

HEADING	Parameter	Units
LAKE	Lake number	as assigned
RIVER	River code	after Waldron (1964)
LAT	Latitude	in degrees, minutes
LONG	Longitude	in degrees, minutes
DATE	Sampling date	year, month, day
ORDER	Drainage order	as assigned
GEOLOGY	Bedrock geology (see below)	as assigned
LAREA	Lake surface area	ha
DAREA	Lake drainage area	ha
RATIO	Ratio of drainage to surface	0.00
	area	
ELEV	Elevation	m above sea level
DISTCST	Distance from the coast	km

Note: Assigned bedrock geology types are as follows: 4 - gneiss, schist and foliated granite

- 6 granite
 7 gabbro, diorite, pyroxenite, granodiorite, and mafic syenites
 9 acidic to mafic volcanic rocks with associated greywacke, siltstone, sandstone, conglomerate, chert, minor limestone, and metamorphic equivalents

Table 1.2 Physical and chemical data for the study lakes (see end of table for an explanation of headings and units).

LAKE TEMP F_PH F_ALK COND HARD TDS

600 601 602 603 604 605 606 607 608 609	16.0 15.5 16.0 17.0 17.0 16.0 15.5 16.0 16.0	6.20 6.02 5.74 5.02 5.56 5.84 5.73 5.40 6.34 5.54	72.8 29.4 24.9 1.4 21.8 29.2 58.2 13.2 45.8 14.5	23.0 13.5 14.3 16.6 11.5 16.2 13.2 11.3 25.0 12.2	9.9 46.0 56.3 49.6 38.6 45.9 55.7 25.3 35.5 34.2	30 10 25 55 55 35 15 40 45 30	3.5 5.0 80.0 100.0 50.0 35.0 100.0 30.0 25.0 50.0	1.5 3.6 3.6 5.4 1.9 2.4 4.2 2.4 3.0 3.1	81 449 435 436 419 347 371 306 318 212	25 27 30 35 30 46 40 34 25 24	39.6 47.9 56.2 56.2 50.0 45.8 35.4 41.6 29.1 33.3
610 611	16.0 16.5	5.34	11.5	9.7 13.2	33.1 63.8	30 20	50.0	3.7	544 266	23 45	33.3 39.6
612	15.0	5.31	-1.5	11.2	23.4	15	15.0	9.0	143	34	29.1
613	16.0	6.18	49.4	30.0	56.2	15	70.0	5.2	281	30	35.4
614	16.0	5.13	0.8	11.0	22.6	15	60.0	3.4	261	23	33.3
615	16.0	5.12	. 3.0	12.8	38.8	20	70.0	3.6	286	26	41.6
616	16.0	5.57	5.6	10.0	26.5	55 15	25.0	9.9	153	20	27.1
617 618	17.0 16.0	5.18 5.09	9.1 -11.4	9.7 9.8	24.9 20.9	15 30	35.0 30.0	2.4 9.3	272 270	24 20	37.5 37.5
619	16.0	5.38	-11.4 -5.4	9.8	22.6	20	60.0	3.2	554	20	27.1
620	16.0	5.92	29.8	9.5	33.8	35	45.0	4.0	397	20	35.4
621	16.0	5.58	14.6	9.3	23.2	30	30.0	2.6	407	42	27.1
622	16.0	5.65	11.9	10.8	32.3	25	25.0	4.3	399	25	29.1
623	15.0	5.52	15.4	8.5	20.6	30	60.0	2.0	383	23	22.9
624	16.0	5.87	10.4	9.0	24.3	40	35.0	2.0	398	20	22.9
625 626	17.0 16.0	5.55 6.17	11.9 34.4	8.8 12.0	21.8 42.1	35 10	50.0 28.0	2.3	275 171	35 48	27.1 27.1
627	16.5	5.33	-1.8	11.9	20.8	15	22.0	0.3	400	55	27.1
628	17.0	5.45	5.9	12.0	37.5	25	70.0	4.3	422	45	27.1
629	17.5	5.51	23.2	11.1	42.0	30	60.0	4.8	314	45	27.1
630	17.0	5.30	-3.3	37.0	19.0	10	20.0	4.1	369	37	27.1
631	17.0	6.36	42.4	9.7	40.7	15	25.0	3.7	367	40	33.3
632	17.0	5.58	8.9	9.3	27.5	30	15.0	2.7	374	43	27.1
633	17.0	6.25	44.8	9.3	41.3	10	30.0	4.0	394	33	22.9
634	17.5 18.0	5.72	15.2 35.5	11.0	26.0	10	27.0 25.0	5.2	378 389	35	39.6 39.6
635 636	19.0	6.43 5.97	36.5	12.8 12.2	41.8 52.3	40 20	60.0	4.0	389 477	30 41	39.6 37 . 5
637	17.5	6.63	52.0	13.8	40.4	45	25.0	3.4	447	32	35.4
638		6.02	15.2	33.0	30.0	15	20.0		390	33	29.1
639	17.0	6.44	27.0		21.1	15	12.0	2.4	211	23	33.3
640	17.0	6.63	62.3	14.0	49.4	20	50.0	2.9	149	33	29.1
641	16.0	5.03	-3.5	9.0	19.8	15	50.0	3.2	424	24	33.3
642	16.5	6.30	10.0	19.5	20.6	20	10.0	3.1	390	40	27.1
643		6.26	17.0	7.5	27.9	10	15.0	3.9	513	20	22.9
644	16.0	4.65	0.0	30.0	19.6	10	15.0	4.0	389	30	33.3

LAKE	TEMP	F_PH	F_ALK	COND	HARD	TDS	COL	DOC	ALUM	MANG	MTBS04
645	17.0	5.67	9.3	9.0	27.4	15	35	3.9	391	30	33.3
646	17.0	6.54	65.2	11.1	61.0	80	25	4.2	383	25	29.1
647	16.0	6.25	16.9	12.1	56.4	35	100	6.3	417	49	29.1
648	16.0	5.59	0.0	45.0	46.5	10	50	5.3	380	45	27.1
649	16.0	6.65	86.6	12.0	75.0	125	30	4.5	403	24	27.1
650	17.0	6.60	4.3	12.0	76.4	95	50	4.8	383	26	33.3
651	17.0	4.90	0.0	22.0	43.0	100	40		392	22	27.1
652	17.0	6.65	154.8	18.3	57.9	115	60	9.3	394	36	27.1
653	17.0	5.68	-2.0	7.5	36.3	130	30	4.7	381	15	22.9
654	18.0	6.12	34.0	8.2	36.8	30	35	4.9	392	21	22.9
655	17.0	5.12	0.6			4.05					0.0
656	17.0	6.06	37.8	9.6	30.3	105	30	4.7	415	19	22.9
657	16.0	5.98	22.8	9.2	26.1	85	40	3.4	412	20 .	18.7
658	17.0	4.80	-9.5	10.8	24.7	120	100	5.3	142	22	27.1
659	17.0	5.34	-0.5	10.0	26.3	115	40	4.7	420	23	20.8
660	17.0	5.71	13.9	9.0	42.2	10	25	4.9	390	46	18.7
661	16.0	5.07	9.9	9.5	50.8	25	100	4.6	395	48	20.8
662	19.0	5.18	5.8	9.9	24.3	10	60	6.1		30	22.9
663	17.0	5.79	29.9	10.0	33.2	180	100	5.9		20	27.1
664	17.0	5.39	14.0	9.0	28.5	135	70	7.1	418	20	27.1
665	18.0	5.17	-5.6	20.0	17.7	30	20	5.6	412	20	16.7
666	18.0	5.58	7.4	9.0	26.6	15	50	3.6	394	16	18.7
667	16.0	5.20	8.9	9.5	19.4	85	30	2.6	404	20	27.1
668	17.0	6.65	64.1	12.0	89.3	20	30	3.6	387	32	18.7
669	18.0	5.20	25.2	12.0	59.3	45	100	3.3	449	28	25.0
670	18.0	4.90	3.3	12.0	21.0	80	80	4.1	413	20	18.7
671	18.0	4.72	-29.6	10.3	22.4	45	100	3.1	454	25	25.0
672	17.0	4.82	-1.2	8.0	24.8	45	50	2.9	216	22	29.1
673	14.0	5.86	0.0	76.0	115.2	10	120	20.0	92	76	29.1
674	17.0	6.64	51.5	12.0	66.6	50	26	10.9	33	28	18.7
675	15.0	6.22	26.7	8.0	40.1	90	30	9.6	372	19	18.7
676	16.0	6.51	93.9	14.0	76.4	140	25	40 5	388	28	31.2
677	16.0	6.12	43.2	13.0	69.2	105	80	13.5	403	25	29.1
678	16.0	5.40	6.4	7.2	22.2	85	15	14.7	378	24	27.1
680	17.0	5.04	2.5	11.0	22.9	100	80	4.7	430	28	29.1
681	16.0	6.13	33.5	11.0	37.4	170	50	8.4	434	22	27.1
682	16.0	6.49	40.0	10.0	52.2	150	50	11.7	59	31	25.0
683	17.5	6.01	45.6	11.5	34.3	80	15	3.3	441	30	37.5
685	16.0	5.98	25.6	14.5	56.3	75	50	2.3	394	26	22.9
686	17.0	4.91	-0.4	10.5	29.2	105	100	4.2	443	24	33.3
687	18.0	4.61	-14.0	25.0	35.0	305	100	3.6	385	25	16.7
688	17.0	6.14	35.9	8.8	40.5	170	50	3.0	399	21	27.1
691	18.0	6.56	53.6	12.6	55.4	30	120	15.0		30	18.7
692	17.0	6.67	49.9	10.5	33.2	35	25		•	35	16.7

Table 1.2 (Cont'd.)

LAKE CALC MAGN SOD POT H ION AL CATIONS HCO3 CHL SULP ORG 0.6 8.9 217.6 88.7 93.1 35.4 14.0 231.2 1.0 49.4 215.5 35.8 101.6 54.1 33.1 224.6 43.5 4.6 600 100.0 60.0 1.8 47.9 236.5 30.3 87.5 56.2 32.0 206.0 9.5 48.0 235.6 1.7 87.5 50.0 41.6 180.8 2.8 46.1 201.6 26.6 124.1 56.2 16.4 223.3 1.4 38.2 229.3 35.6 166.4 60.4 21.6 284.0 1.9 40.8 230.2 70.9 110.0 43.7 37.3 261 9 40.0 33.7 157.5 35.8 101.6 54.1 33.1 224.6 30.3 87.5 56.2 32.0 206.0 1.7 87.5 50.0 41.6 180.8 601 49.9 25.7 82.6 7.0 87.0 5.8 95.7 2.9 64.9 29.1 602 603 48.7 30.8 34.9 25.7 604 87.0 5.2 41.2 30.8 113.1 4.6 59.9 31.4 91.3 4.9 605 606 22.5 17.1 73.9 3.5 4.0 33.7 154.6 16.1 95.9 56.2 20.2 188.4 607 0.5 35.0 173.7 55.8 110.0 39.6 28.4 233.8 608 38.7 19.7 76.1 3.8 2.9 23.3 161.6 17.7 101.6 47.9 26.7 193.9 609 26.2 25.7 78.3 5.2 14.0 87.5 43.7 30.7 175.9 610 32.4 20.6 69.6 3.2 4.6 59.8 190.2 89.8 23.1 82.6 3.8 0.4 29.3 229.0 85.6 87.5 47.9 34.2 255.2 611 4.9 15.7 136.0 0.0 101.6 37.5 74.2 213.3 18.7 17.1 76.1 3.5 612 0.7 30.9 215.8 60.2 81.8 37.5 48.6 228.1 613 67.4 27.4 84.8 4.6 7.4 28.7 140.8 62.1 33.3 26.9 123.3 614 20.0 15.4 65.2 4.1 1.0 7.6 31.5 177.7 615 37.4 24.5 73.9 2.9 3.7 76.2 47.9 28.5 156.3 25.0 17.1 65.2 4.6 2.7 16.8 131.4 6.8 81.8 37.5 85.7 211.8 616 617 23.7 15.9 73.9 3.2 6.6 29.9 153.2 11.1 79.0 47.9 19.2 157.2 16.5 15.4 69.6 3.5 8.1 29.7 142.8 0.0 81.5 62.5 73.0 217.0 618 20.0 15.4 80.5 2.9 4.2 60.9 183.9 0.0 87.5 47.9 26.8 162.2 619 36.2 19.2 1.2 43.7 177.0 76.2 27.1 36.4 176.0 620 71.8 4.9 36.3 140.9 17.8 84.6 27.1 22.5 152.0 621 22.5 14.6 52.2 4.2 2.6 44.8 81.8 31.2 37.7 165.2 53.6 20.8 17.2 110.4 59.2 16.7 18.1 106.7 69.6 3.5 54.4 4.1 38.7 15.8 622 173.7 14.5 2.2 43.9 137.6 148.8 22.5 11.5 3.0 42.1 18.8 623 60.9 2.9 624 26.2 13.7 1.3 43.8 12.7 14.5 73.3 35.4 19.8 143.0 41.9 73.3 31.2 19.6 166.0 0.0 101.6 31.2 2.5 135.3 7.2 81.8 31.2 36.4 156.6 28.3 70.5 27.1 41.1 167.0 22.5 12.9 65.2 3.5 52.4 19.4 69.6 3.5 137.2 625 2.8 30.3 626 0.7 18.8 164.4 16.2 15.4 67.4 3.8 4.7 44.0 151.5 627 628 39.9 21.4 73.9 2.6 3.5 46.4 187.8 47.4 22.3 69.6 2.0 3.1 34.5 178.9 629 79.0 31.2 33.7 143.9 84.6 35.4 35.1 206.7 84.6 27.1 23.4 145.9 67.7 22.9 37.6 182.8 15.5 13.7 44.9 22.3 5.0 40.6 131.1 0.0 52.2 4.1 630 185.7 157.8 73.9 3.8 0.4 40.4 51.6 631 27.4 16.8 65.2 4.6 2.6 41.1 10.8 632 0.6 43.3 178.8 54.6 47.4 21.4 60,9 5.2 633 79.0 37.5 136.0 81.8 43.7 49.5 218.2 22.5 17.9 1.9 41.6 148.7 18.5 60.9 3.9 634 49.9 20.6 0.4 42.8 196.1 78.3 4.1 43.2 635 79.0 39.6 36.6 199.7 44.5 69.6 2.9 214.1 62.4 25.7 1.1 52.5 636 76.2 37.5 32.8 209.8 79.0 33.3 18.4 149.2 63.3 49.9 18.9 65.2 3.5 0.2 49.2 186.9 637 145.5 22.5 22.8 1.0 42.9 18.5 638 52.2 4.1 76.2 33.3 22.9 165.3 70.5 29.1 27.9 203.4 32.9 639 22.5 12.0 50.9 3.8 0.4 23.2 112.8 640 52.4 28.3 60.9 4.1 0.2 16.4 162.3 75.9 56.5 3.2 0.0 70.5 41.6 24.7 136.8 15.7 14.6 9.3 46.6 146.0 641 18.7 13.7 0.5 42.9 129.3 12.2 70.5 27.1 29.2 139.0 642 50.0 3.5 0.5 56.4 159.9 20.7 70.5 22.9 36.7 150.8 32.4 14.2 52.2 4.1 643 22.4 42.8 157.2 3.3 79.0 37.5 27.7 147.5 58.8 4.0 13.6 15.6 644

Table 1.2 (Cont'd.)
SOUTH COAST LAKES - CHEMICAL DATA 2

LAKE	CALC	MAGN	SOD	POT	H_ION	AL	CATIONS	HCO3	CHL	SULP	ORG	ANIONS
645	21.5	20.2	64.0	2.2	2.1	43.0	153.0	11.3	70.5	33.3	34.3	149.4
646	68.0	32.8	61.4	6.3	0.3	42.1	210.9	79.4	59.2	25.0	40.3	203.9
647			85.7			45.9	237.1		73.3		59.2	186.4
648			67.8			41.8	196.3	53.5	79.0,	25.0		203.5
649	104.3					44.3	229.7	105.5	59.2	25.0		233.1
650			58.8			42.1	233.5		62.1		46.2	146.8
651			49.9		12.6		185.1		62.1			101.7*
652			57.6			43.3	193.1	188.5				361.1
653			59.9				168.6		53.6			120.0
654	43.1	18.6	61.4	4.0		43.1	171.0		67.7	22.9	45.5	177.5
655 656	20 E	10 0	49.1	1 /	7.6	45.7	145.4	0.7	70.5	25 0	/2 /	107.0
657			65.2		1.0							184.9 143.9
658			56.3		15.8		130.0	0.0	50 8	31 2	38 4	120.4
659			60.1	2 5	4.6	46 2	154.8	0.0	64.9	25.0	39.0	128.9
660			64.0	2.3	1.9	42.9	156.8 130.7 154.8 179.4	16.9	87.5	22.9	43.3	170.6
661			61.4	2.6	8.5	43.5	196.4	12.1	64.9	25.0	35.9	137.9
662			61.3		6.6		124.3*	7.1				151.5
663			57.8		1.6		134.7*		50.8			160.8
664	22.7	20.9	57.8	2.5	4.1	46.0	154.0	17.1	62.1	22.9	59.5	161.6
665			61.4			45.3	142.6	0.0				154.4
666			48.6			43.3	139.5		50.8			105.6
667			53.7			44.4	137.6		70.5			125.1
668			48.6			42.6	235.3		39.5			164.8
669			69.1		6.3		226.4	30.7	64.9	18.7	26.6	140.9
670			58.3		12.6		150.7	4.0	64.9	22.9	30.6	122.4
671			64.0		19.1				59.2 59.2			104.0
672 673	158.8		60.1			23.8	142.3 292.1				180.5	107.4
674	79.4						171.8				105.1	
675			46.0			40.9	151.0		53.6			192.8
676			64.0			42.7	238.6	114.4				222.5*
677			76.7			44.3	246.1				125.4	
678			40.4		4.0		121.7	7.8	56.4	33.3	123.4	220.9
680	15.9	18.2	71.6	2.5	9.1	47.3	164.6	3.0	67.7	25.0	36.4	132.1
681	45.4	17.9	64.0	4.7		47.7	180.5	40.8	84.6	22.9	78.1	226.4
682			58.8			6.5	153.6				111.8	
683			92.1			48.5	202.1		95.9			223.3
685			83.1			43.3	225.9		81.8			157.0
686			56.3			48.7	166.6		59.2			117.7
687			30.7			42.4	115.6		50.8			87.9
688			58.8			43.9	168.7		48.0			142.5
691			54.8		0.3		132.2*				143.9	
692	/.5	33.9	45.2	3.6	0.2		112.4*	8.00	53.6	22.9		137.3*

Table 1.2 (Cont'd.)

LAKE	XCALC	XMAGN	XSOD	XPOT	XSULP	PHOS	NITR	IONB	IONB1
600	98.0	53.8	0.0	2.8	22.5	0.000	0.032	-2.1	-3.0
601	47.8	18.9	26.2	4.9	40.0	0.076	0.032	-7.4	-2.1
602	63.0	23.3	38.4	4.1	44.1	0.001	0.032	3.5	6.9
603	46.8	25.0	47.1	1.2	37.8	0.001	0.032	12.3	13.2
604	32.3	17.4	18.0	2.7	39.0	0.001	0.032	-15.0	-5.1
605	37.7	19.7	20.5	1.3	37.2	0.001	0.032	-16.1	-10.7
606 607	57.6 20.4	24.0 10.7	30.2 20.6	2.7	28.4	0.184	0.032	-9.0	-6.4
608	36.4	12.3	14.9	1.6 1.6	42.9 24.3	0.001	0.032	-18.0 -19.5	-9.8 -14.7
609	24.1	18.9	21.8	3.2	33.8	0.001	0.032	-19.5	-14.7 -9.1
610	30.6	14.7	21.0	1.4	31.6	0.001	0.032	-7.2	3.9
611	88.0	17.3	34.0	2.0	35.7	0.001	0.032	-5.2	-5.4
612	16.6	10.3	19.6	1.5	23.4	0.000	0.032	-9.3	-22.1
613	65.6	21.9	39.3	3.0	26.1	0.003	0.032	1.3	-2.8
614	18.7	11.3	30.7	2.8	24.7	0.001	0.032	4.1	6.6
615	35.8	19.4	31.6	1.4	37.3	0.002	0.032	4.1	6.4
616	23.2	11.7	19.7	3.0	26.1	0.001	0.032	-6.0	-23.4
617	22.0	10.6	30.0	1.6	36.9	0.002	0.032	-8.4	-1.3
618	14.8	9.9	24.3	1.9	44.4	0.001	0.032	-15.7	-20.6
619	18.1	9.6	31.8	1.2	35.7	0.002	0.032	-6.5	6.3
620	34.6	14.1	29.4	3.4	16.5	0.001	0.032	-2.8	0.3
621	20.7	8.9	5.1	2.5	15.3	0.001	0.032	-16.1	-3.8
622	37.0	10.3	24.1	1.9	19.9	0.001	0.032	0.0	2.5
623	21.3	7.9	24.6	3.0	13.4	0.002	0.032	-0.4	11.0
624	25.0	9.7	27.9	1.7	8.4	0.002	0.032	7.9	16.5
625	20.9	7.9	24.5	2.0	25.2	0.001	0.032	-8.4	-2.1
626	50.9	14.5	28.8	2.0	21.0	0.001	0.032	-0.5	-0.5
627	14.1	8.6	10.9	1.7	17.1	0.001	0.032	-12.7	5.6
628	38.2	15.9	28.4	1.0	19.9	0.001	0.032	6.8	9.0
629 630	45.9 13.8	17.6 8.4	30.4 8.3	0.6	17.3 20.3	0.001	0.032	5.8 -12.6	3.4
631	43.1	16.6	26.9	2.5 2.1	23.6	0.001	0.032	-12.6 -8.4	-4.7 -5.3
632	25.7	11.1	18.2	3.0	15.3	0.012	0.032	-3.6	3.9
633	46.0	16.9	23.2	3.9	13.5	0.001	0.032	-3.7	-1.1
634	20.8	12.6	17.0	2.3	26.5	0.000	0.032	-12.4	1.1
635	48.2	15.1	32.8	2.4	32.4	0.000	0.032	-4.9	~5.3
636	60.7	20.4			28.6	0.001		-0.8	
637	48.3	13.7	22.9	2.0	26.9	0.000	0.032	-12.6	-5.8
638	20.8	17.5	8.3	2.5	22.3	0.103	0.032	-12.6	-1.3
639	20.9	6.9	8.5	2.3	22.7	0.023	0.032	-23.0	-18.9
640	50.9	23.6	21.7	2.7	19.3	0.001	0.032	-9.3	-11.2
641	14.2	9.8	17.3	1.8	31.8	0.126	0.032	-10,9	3.2
642	17.2	9.0	10.8	2.1	17.3	0.001	0.032	-12.2	-3.6
643	31.0	9.5	13.0	2.7	13.1	0.001	0.032	-5.2	2.9
644	12.0	10.3	14.9	2.4	26.5	0.001	0.032	-13.1	3.2

Table 1.2 (Cont'd.)

LAKE	XCALC	XMAGN	XSOD	XPOT	XSULP	PHOS	NITR	IONB	IONB1
645	20.1	15.5	24.7	0.8	23.5	0.001	0.032	-3.2	1.2
646	66.8	28.9	28.5	5.1	16.7	0.001	0.032	1.5	1.7
647	71.0	19.6	44.9	6.5	23.1	0.001	0.032	20.0	12.0
648	52.8	18.3	23.9	4.5	14.0	0.002	0.032	-1.8	-1.8
649	103.1	23.9	16.9	1.8	16.7	0.001	0.032	-1.2	-0.7
650	91.7	32.3							
			24.3	1.6	24.7	0.001	0.032	31.0	22.8
651	60.6	10.6	15.4	1.6	14.3	0.001	0.032	12.0	00.0
652	47.0	37.0	23.1	1.3	12.2	0.002	0.032	-29.0	-30.3
653	44.2	13.0	30.1	1.6	17.5	0.002	0.032	22.6	16.8
654	41.7	14.0	23.8	2.6	13.5	0.001	0.032	-1.9	-1.9
655				0.0		0.000			
656	28.0	14.2	9.9	0.0	15.2	0.001	0.032	-17.7	-11.9
657	23.6	12.4	30.7	2.4	14.3	0.001	0.032	-1.1	4.3
658	23.9	11.5	28.0	1.9	24.2	0.002	0.032	9.5	4.1
659	22.5	13.2	24.0	1.2	16.0	0.001	0.032	7.3	9.1
660	41.3	19.3	15.3	0.6	10.7	0.002	0.032	2.8	2.5
661	46.3	28.5	25.3	1.3	16.0	0.001	0.032	17.2	17.5
662	28.5	22.9	22.1	1.4	15.2	0.012	0.032	8.5	
663	31.9	34.5	29.6	3.0	13.8	0.029	0.032	10.4	
664	21.4	16.7	23.3	1.3	14.3	0.001	0.032	0.9	-2.4
665	9.6	8.9	14.3	1.5	13.2	0.086	0.032	-9.5	-4.0
666	21.6	15.2	20.4	2.6	7.5	0.002	0.032	11.4	13.8
667	15.5	8.5	14.5	1.5	13.1	0.017	0.032	-9.1	4.8
668	80.8	56.4	26.6	2.5	7.0	0.017	0.032	19.3	17.6
669	64.4	27.8	33.0	2.3	9.7	0.002		19.3	
		10.9	22.3	0.8	13.9	0.002	0.032	0.5	23.3
670	15.6						0.032		10.4
671	15.3	13.3	31.0	1.3	14.7	0.002	0.032	10.0	23.9
672	23.7	11.1	27.2	2.1	18.8	0.001	0.032	9.0	14.0
673	157.1	38.5	30.3	2.2	14.0	0.001	0.032	10.4	-16.6
674	78.0	28.3	13.5	3.1	9.3	0.000	0.032	5.9	-19.3
675		28.3	16.2	2.7	9.2	0.001	0.032	3.2	-12.1
676	89.1	32.5	20.0	1.5	18.2	0.001	0.032	-6.4	
677	82.1	27.5	29.7	5.5	23.6	0.001	0.032	7.6	-9.5
678	19.2	10.8	9.1	0.0	25.5	0.001	0.032	-12.3	-29.0
680	14.5	13.6	34.0	1.2	15.6	0.003	0.032	6.1	11.0
681	43.6	12.2	16.9	3.0	11.1	0.001	0.032	-5.8	-11.3
682	50.6	26.9	18.1	2.5	12.7	0.001	0.032	0.7	-25.1
683	32.0	14.7	38.8	3.5	28.3	0.002	0.032	-11.7	~5.0
685	66.3	21.7	37.7	1.6	11.5	0.009	0.032	14.4	18.0
686	26.0	15.1	23.3	1.9	18.8	0.002	0.032	10.1	17.2
687	5.7	6.9	2.5	0.0	5.4	0.005	0.032	-13.0	13.6
688	35.3	24.0	32.2	0.8	16.2	0.001	0.032	4.0	8.4
691	10.4	56.5	23.4	4.1	13.0	0.001	0.032	-3.9	- · ·,
692	6.4	32.3	15.4	2.5	15.5	0.001	0.032	-19.7	
V / 4	J • 7	J- • J	~~.~			0.002	J . J J =		

Table 1.2 (Cont'd.)

Key to parameter names and units of expression

HEADING	Parameter	Units
LAKE TEMP F_PH F_ALK COND HARD TDS COL DOC ALUM MANG MTBSO4 CALC MAGN SOD POT H_ION AL CATIONS HCO3	Lake number Water temperature pH (meas. in field) alkalinity (meas. in field) Conductivity Hardness Total dissolved solids Water colour Dissolved organic carbon Aluminum Manganese M.T.B. sulphate Calcium Magnesium Sodium Potassium Hydrogen ion Aluminum Sum of cations Bicarbonate Chloride I.C. sulphate Organic anions Sum of anions 'Excess' calcium 'Excess' magnesium 'Excess' sodium 'Excess' sodium 'Excess' solium 'Excess' sulphate (I.C.) Ortho-phosphate	Units as assigned C pH units µeqL-¹ µScm-¹ µeqL-¹(CaCO3) mgL-¹ µgL-¹ µgL-¹ µeqL-¹ µeqL-¹
NITR IONB IONB1	Nitrate Ion balance (see below) Ion balance (see below)	µeqL-¹ % %

Note: IONB includes the cations calcium, magnesium, sodium, potassium, and the anions bicarbonate, chloride, and sulphate(I.C.). IONB1 includes IONB and the additional cations, hydrogen ion and aluminum as well as the anion, organic anions.

