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THE EFFECT OF HALIFAX AIR EMISSIONS ON THE WATER QUALITY OF THIRTY-SIX HALIFAX AREA LAKES, 1980

Surveillance Report EPS-5-AR-84-2
Atlantic Region

Canada

ENVIRONMENTAL PROTECTION SERVICE REPORT SERIES

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THE EFFECT OF HALIFAX AIR EMISSIONS ON THE WATER QUALITY OF THIRTY-SIX HALIFAX AREA LAKES, 1980

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February, 1983

ABSTRACT

Water samples were collected on June 3, 1980 from thirty-seven undisturbed lakes within 10 to 70 km of Halifax, Nova Scotia to evaluate the impact of urban air emissions on lake water quality. These lakes lie on granitic or occasionally quartzitic rock. As a group they were dilute and acidic, with mean Gran alkalinities of -7 to -15 μeq/l and mean pH values of 4.9 to 4.7, west and east of Halifax, respectively. The results suggest that the influence of local acid producing emissions is restricted to areas <30 km from the city and is significant only east of Halifax. In the east the results suggest an increase of 59 μeq/l (2.8 ppm) in excess sulphate, i.e., a doubling of normal lake concentrations. This is accompanied by a significant increase in lake acidity of 11 μeq/l, which results in a small (approximately 0.2 units) but not significant decline in lake pH. This generally confirms previously reported results and documents measurable effects of urban sulphur emissions on adjacent lake water quality.

RÉSUMÉ

Des échantillons d'eau de trente-sept lacs non perturbés, dans un rayon de 10 à 70 km d'Halifax (Nouvelle-Écosse) ont été prélevés le 3 juin 1980; ce travail servait à évaluer l'incidence des émissions gazeuses en provenance des villes sur la qualité des eaux lacustres. Les lacs recouvrent des granites et occasionnellement des quartzites, et considérés en groupe, ont des eaux diluées et acides, une alcalinité moyenne de Gran de -7 à -15 μeq/l, et un pH moyen oscillant entre 4.9 et 3.7, valeurs mesurées à l'ouest et à l'est d'Halifax respectivement. Les résultats suggèrent que les émissions acides locales n'exercent d'influence qu'à moins de 30 km d'Halifax, et ne sont significatives qu'à l'est de cette ville. A l'est, parmi les effets suggérés, on indique une augmentation de 59 μeq/l (2.8 ppm) du sulfate excédentaire dont la concentration passe alors au double de la normale pour les eaux lacustres. En même temps, celles-ci manifestent une nette augmentation d'acidité, qui entraîne une diminution légère (d'environ 0.2 unités) et peu significative du pH lacustre. Ces faits confirment généralement les résultats établis, qui nous donnent une mesure de l'incidence des émissions sulfureuses d'origine urbaine sur la qualité des eaux lacustres proches.

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1. INTRODUCTION

On June 3, 1980, a water quality survey by helicopter was conducted of 39 lakes within 10 to 75 km of Halifax, Nova Scotia (Figure 1). The purpose of the study was to evaluate the influence on the terrestrial and aquatic receiving environment of the air emissions from Halifax, particularly those originating from power generation. The study was divided into two portions: lake water quality sampling, and the monitoring of trace metals in vegetation. This report presents the findings of the aquatic portion of the study.

Of the study lakes, 18 were located to the east of the City and 21 were located to the west (Appendix Table 1). These lakes were selected based on their lack of development and their common bedrock geology. With few exceptions, the lakes sampled are not accessible by road and the direct influence of man is minimal. The sites chosen are representative of granitic rocks of lower or middle Devonian origin, with four exceptions (Lakes 14, 16, 42 and 45) which are on metamorphic strata. This was done in an attempt to reduce the influence of variable geology on the water chemistry results.

A number of studies have documented the effects of point sources of SO_2 on adjacent lake chemistry (Gorham and Gordon, 1960; Conroy, 1974; and Yan and Stokes, 1976). Effects observed in these studies include reduced lake pH, elevated lake sulphate levels, and reduced buffering capacity. Published studies of lakes near Halifax are limited and include Gorham (1957), and Watt et al. (1979). Watt et al. (1979) reports a significant pH decline in 16 lakes, within 40 km of Halifax during a 21 year period. He also suggests that non-marine sulphate and hydrogen ion

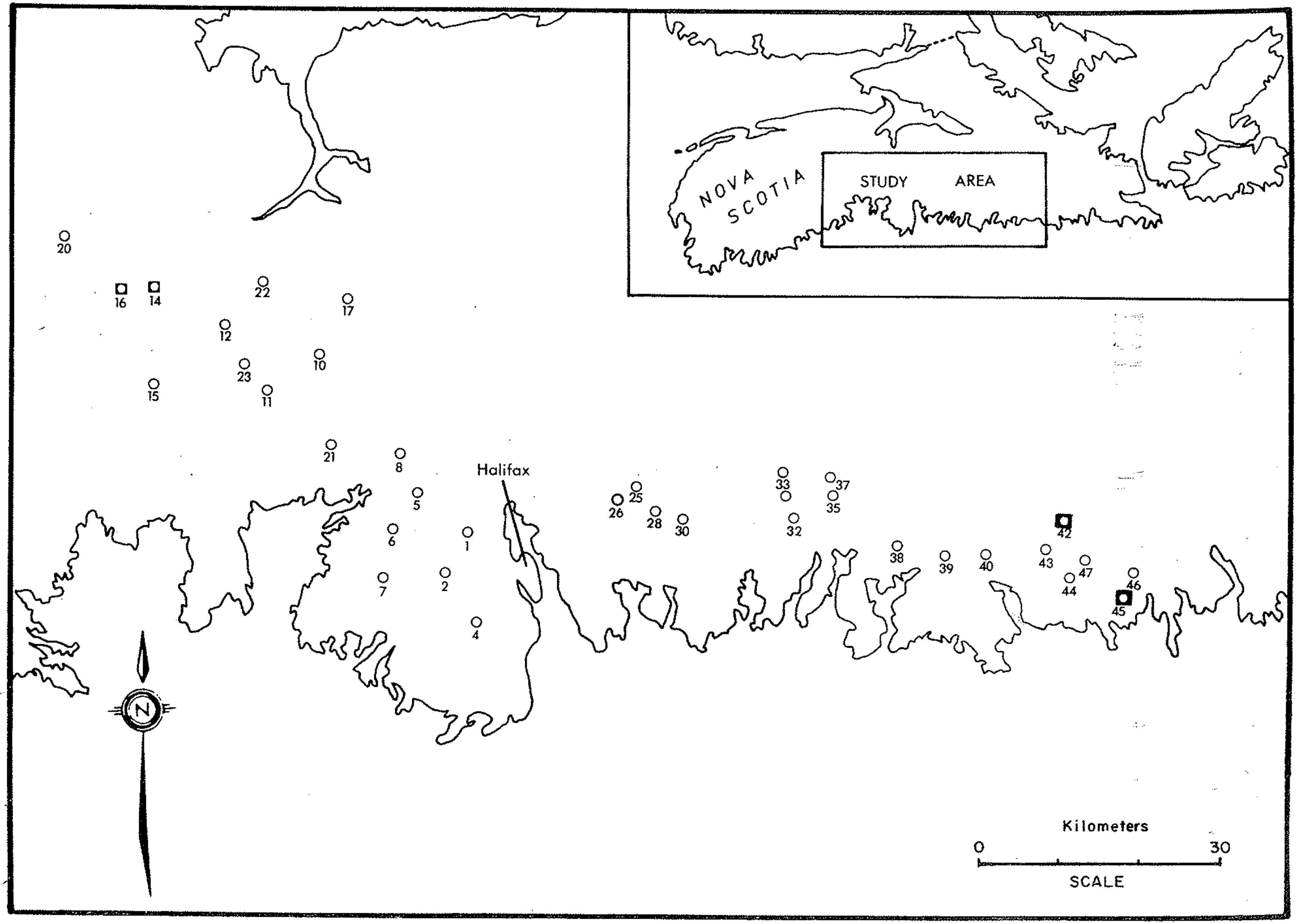

FIGURE 1 : THE LOCATION OF THE 36 STUDY LAKES IN THE HALIFAX AREA OF NOVA SCOTIA, CANADA.

concentrations in lakes are significantly correlated with distance from Halifax and concludes that the cause is local air emission sources. Shaw (1982) estimates that 50% of the annual wet deposition of hydrogen ion and sulphate at St. Margaret's Bay, a site 25 km west of Halifax, is due to emissions from the City. This deposition, carried by easterly winds from Halifax, is estimated as 36 kg $SO_4^=$ ha^{-1} and 43 meq H^+ m^{-2} per year. Principal SO_2 producing sites in the urban area include two oil fired generating stations and two oil refineries. Total metropolitan emissions are estimated at 1800 g SO_2 s^{-1} (Shaw, 1982).

2. METHODS

Water samples were collected during a single six hour period on June 3, 1980. Samples were collected with a two liter Kemmerer PVC sampler from a hovering helicopter. A single two-liter sample was obtained from each lake at a depth of 0.5 to 1.5 meters, transferred to an acid washed, deionized water rinsed polythylene bottle and the temperature recorded. The samples were kept cool and were delivered to the laboratory within 3 hours of collection. In the laboratory two subsamples were created: one for pH and alkalinity analysis, and one (250 ml) for trace metal analysis which was preserved with 2 ml of reagent grade HNO_3. Using a Radiometer PHM64 pH meter and a combination glass electrode (Radiometer GK2401C), pH measurements were performed on the appropriate sub-samples immediately. Total alkalinity was determined potentiometrically by titration and the equivalance point was determined from the intercept of a Gran plot (Stumm and Morgan, 1970). A more complete description of these procedures and the other chemical methods used by the EPS laboratory for the analysis of the samples are given in Wiltshire and Machell (1981). Estimates of marine and excess levels of [SO_4], [Ca] and [Mg] were based on measured levels of chloride ion in the samples.

Peculiarly low pH results occurred in three of the lake samples. A subsequent resampling of one of these three in October, 1980, did not confirm the earlier results and the data for the three have been eliminated. In the remaining 36 lakes, confusion exists over the source of samples 25 and 26 as to which is from Duddley and which is from East Lake. As the distance from Halifax for both is so similar, the data has been used in the report.

Regression equations for pH, [Ca] + [Mg], and Gran alkalinity were calculated using the expression $C = a+b \log X$, where C is the concentration of the parameter at a distance X km from Halifax. Excess sulphate data were normalized by log transformation. This relationship assumes that beyond the distance of maximum deposition (<10 km), lake concentration should decrease with increasing distance from Halifax, if Halifax sources are significant contributors. The sum of calcium and magnesium was regressed to test the expectation that major changes of geology with distance did not influence the alkalinity data. The regressions were tested for non-zero slope using the Student statistic calculated, using the variance of X and Y and the mean square deviation of sample points from the estimated regression line. The number of degrees of freedom was N-2 with N the number of samples.

3. RESULTS AND DISCUSSION

3.1 Ion Balances

Balances of the cations and anions were calculated for each sample. The ratio of cations to anions ions was 0.9 to 1.1 for 28.6% (10) of 35 samples. The remaining 25 samples or 71.4% of the total had ratios outside this range. Of these, a total of 16 or 45.7% of the total number of samples had ratios <0.8 or >1.2. In this latter group, the poor ion balances can be explained for a number of the samples by unexplicably high sulphate or chloride values. These values were not used in further analysis of the data. These latter ratios are not characteristic of the normal analysis of local lake water. In results of an April, 1980, metropolitan area lake sampling project, whose analyses were performed by another local laboratory, 85.3% of the ion ratios ranged from 0.9 to 1.1 (81 of 95 samples) and no samples had ratios of <0.8 or >1.1 (Gordon *et al*., 1981).

3.2 Water Chemistry

Of the 36 lakes sampled, 18 ranged from 13.5 to 76.8 km east of Halifax, and the remainder were located from 10.4 to 74.3 km west of Halifax. Locations of the lakes are shown in Figure 1 and descriptions are given in Appendix Table 1. The study lakes have been treated as two separate groups, those to the east of Halifax and those to the west. This was done based on the assumption that the influence of Halifax emissions will be more pronounced in the east due to the direction of the prevailing winds (Appendix Figure 1). The wind rose for Shearwater indicates that on the average winds originate between NW and SW 41% of the time, while winds originating from NE to SE occur only 15% of the time.

The water quality results are given in Appendix Table 2, and a summary of these are shown in Table 1. The two groups of lakes, east and west of Halifax, have a very similar water chemistry reflecting their common, primarily granitic geology. Mean pH* values for the two groups are 4.87 and 4.72 while mean Gran alkalinities are -7 and -15 µeq/l. These lakes then can be generally described as acidic with no bicarbonate buffering capacity. Mean levels of sodium and chloride in the east were 126 µeq/l and 128 µeq/l respectively and 140 and 113 µeq/l respectively in the west. Sodium and chloride ratios averaged 0.64 (sea water = 0.56) and ranged from 0.39-0.93. This ratio is close to ratios reported for precipitation (0.63-0.59), and both indicate a slight excess of sodium (Hart, 1977). This mean ratio is consistent with a mean of 0.65 reported by Gorham (1957) and 0.56 reported by Watt _et al_. (1979) for Halifax lakes, as well as those reported by Kerekes (1980), 0.69-0.55, for lakes on granite remote from Halifax. These lakes are heavily influenced by sea salt with sodium and chloride as the dominant cation and anion respectively. If this marine contribution is subtracted, hydrogen ion is usually the dominant cation and sulphate becomes the principal anion. Mean excess sulphate levels were 63 and 62 µeq/l in the west and east respectively, while individual values ranged from 20 to 115 µeq/l. Excess calcium values in the east averaged 23 µeq/l and averaged 34 µeq/l in the west while excess magnesium averaged 22 µeq/l in both directions. Calcium and magnesium concentrations are low which contributes to the low buffering capacity and hence acidity of these waters.

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*$\mathrm{pH} = -\log\left(\frac{\Sigma 10^{-\mathrm{pH}}}{N}\right)$

TABLE 1 - A SUMMARY OF ANALYTICAL DATA FOR THE STUDY LAKES EAST AND WEST OF HALIFAX, JUNE 3, 1980

Parameter	East			West		
	N	$\overline{X}$	Range	N	$\overline{X}$	Range
pH $\left(\frac{-\log [H^+]}{N}\right)$	18	4.72	4.38-5.67	18	4.87	4.37-5.66
Gran Alkalinity µeq/l	18	-15	-44-+8	18	-7	-42-+17
Excess Sulphate µeq/l	14	63	20-105	17	62	30-115
Excess Ca µeq/l	17	23	12-58	18	34	18-70
Excess Mg µeq/l	17	22	15-33	18	22	13-22
Cl^- µeq/l	18	128	87-178	18	140	79-496
Na µeq/l	18	126	104-157	18	113	87-196
K µeq/l	18	9	7-11	18	7	<64-19
Color (units) Median	18	40-50	10-100	18	40	5-100
TOC mg/l	18	6.7	2-10	18	61	2-10
Fe ppb	17	111	40-220	18	111	10-260
Al ppb	17	240	150-380	18	190	40-340
Mn ppb	17	30	10-60	17	30	<10-100
Cu ppb	18	<10	<10	18	<10	<10
Zn ppb	18	<10	<10-30	18	<10	<10
Pb ppb	18	<20	<20	18	<20	<20

The apparent color of the study lakes covered a wide range with 23% less than 20 units (clear), 61.5% between 20 to 50 units and 15% greater than 80 units. Total organic carbon levels ranged from 2 mg/l to 10 mg/l and averaged 6.7 to 6.1 mg/l. A good correlation ($r^2 = 0.76$) of TOC with color was observed (Appendix Figure 2).

The trace metals, [Cu], [Zn] and [Pb] were below detection limits. Total iron levels ranged from 10 to 200 ppb with a mean of 111 ppb. Total aluminum levels ranged from 40 to 300 ppb with means of 240 ppb in the east and 190 ppb in the west. Aluminum levels show a tendency to increase with pH as shown in Appendix Figure 3. Manganese averaged 30 ppb and ranged from <10 to 100 ppb.

To summarize, mean pH and alkalinity levels were slightly lower in the group of lakes to the east of Halifax. Mean excess sulphate and magnesium levels are essentially the same in both directions, while mean excess calcium levels are slightly lower in the eastern group of lakes.

3.3 Acidification

An evaluation of the acidification of the study lakes was made using plots of pH vs excess [Ca] and by calculation using Henriksen's empirical equations (Henriksen, 1979). Henriksen's curve as shown in Figure 2 was developed to distinguish between regions of Norway where the pH of precipitation was <4.6 and >4.6 and where the lakes accordingly have been found to be acidified and non-acidified. Henriksen has demonstrated the curve's applicability to areas of North America for use in identifying acidified lakes. The empirical equation is used to calculate [ACID]

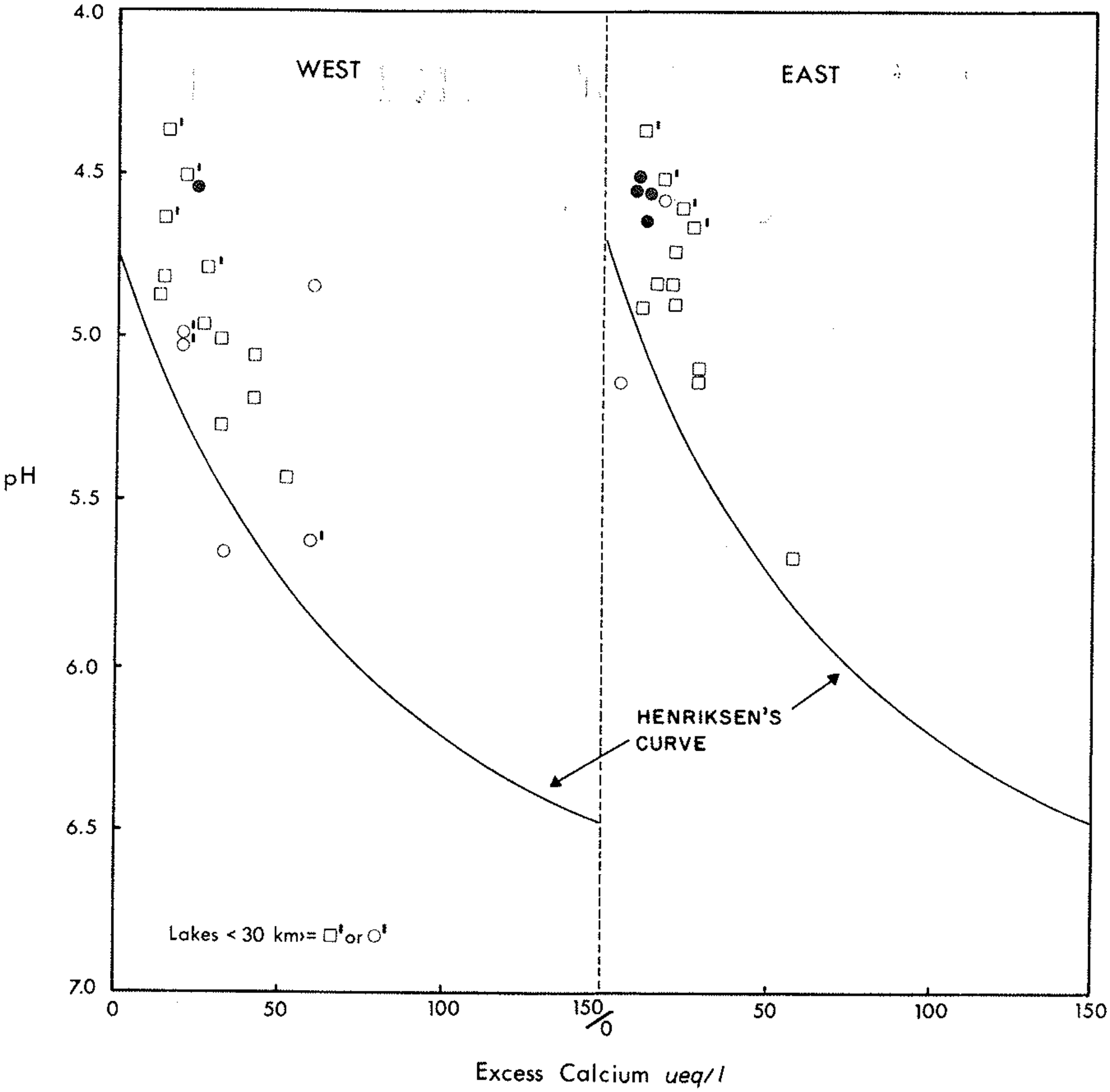

FIGURE 2 pH AND CALCIUM ION CONCENTRATIONS IN HALIFAX AREA LAKES WITH COLOR < 20 (○), COLOR 20–80 (□), AND COLOR > 80 (●). 'HENRIKSEN'S CURVE' FROM HENRIKSEN (1979)

Ca in Table 2 and Appendix Table 3. It is based on the premise that preacidification pH and alkalinity can be estimated from current calcium ion concentrations and that calcium levels remain constant with time. Acidification then by definition involves only a change in alkalinity and assumes a consistent relationship between pH and alkalinity (Henriksen, 1979; Wiltshire and Machell, 1981). The second equation used in Table 2 to calculate [ACID] SO_4 assumes that excess sulphate in the lakes is linearly related to hydrogen ion and excess sulphate in precipitation and that these are related to each other (Henriksen, 1979).

The study lakes east and west of Halifax are plotted for comparison with Henriksen's curve in Figure 2. These plots show that with the exception of two lakes, all fall in the region which is considered acidified, above Henriksen's curve. On inspection, some correlation of position with color is suggested as the plots of the highly colored (>80) lakes are well above the curve. On the basis of this empirical curve (pH vs Ca), lakes within 30 km of Halifax tend to show greater acidification. This tendency is particularly pronounced in the east where four lakes show the highest levels of acidification (Figure 2). This tendency is not explained by high levels of organic material (TOC as indicated by color) as these lakes do not include the lakes with greatest color. Of the eleven lakes within 30 km, five are clear (<20), a total of nine are below 50 units and none is greater than 70 units.

The results of the calculations of the empirical equations are shown in Table 2. The calculation of [ACID] Ca and [ACID] SO_4 are not in agreement, the former being

TABLE 2 - ESTIMATES OF ACIDIFICATION IN THE HALIFAX AREA LAKES ON THE BASIS OF HENRIKSEN'S EMPIRICAL RELATIONSHIPS*

Distance Km	EAST			WEST			ALL		
	N	[ACID] Ca[1]	[ACID] SO_4[2]	N	[ACID] Ca	[ACID] SO_4	N	[ACID] Ca	[ACID] SO_4
Present Study									
0-30	4	28±8	77+10	7	24±15	58±22	11	25±13	65±21
30-50	6	14±7	34±18	5	14±16	45±7	11	22±13	39±15
50-80	8-5	18±13	48±50	6	23±23	36±14	14-11	15±18	41±34
Wiltshire & Machell, 1981									
<30		-	-		-	-	4	53±11	72±27
Gorham, 1957		-	-		-	-	13	-	80±27
Watt, _et_ _al_., 1979		-	-		-	-	13	-	95±21
<40									

* All concentrations are in μeq/1.

[1] [ACID] Ca = -29+1.32 [Ca] - $[ALK]_{80}$; where $[ALK]_{80}$ = Gran alkalinity (μeq/l) for 1980.

[2] [ACID] SO_4 = -12+0.89 [SO_4].

consistently and substantially lower. In earlier lake studies (Wiltshire and Machell, 1981), excellent agreement between these estimates were obtained. The reason for this discrepancy is unknown, and the possibility that the present calcium values are low was considered. For lakes which are in common, the present calcium values agree with those reported by Gordon et al. (1981), but data of both these studies appear to be slightly lower than those reported by Watt et al. (1979). In the single common case, the present excess [Ca] data is lower than Watt's by 18 µeq/l (0.36 mg/l). Such a difference can be explained by yearly and seasonal variation. This difference in excess [Ca] would raise [ACID] Ca estimates (i.e., in Sprucehill Lake) by only 27% and will not account for the magnitude of difference from the reported [ACID] SO_4 estimates. Estimates of average [ACID] SO_4 for all lakes range from 77±10 to 39±15 µeq/l. The mean values do not tend to be higher in the lakes to the east than in those to the west of Halifax (50 vs 49 µeq/l), though the highest estimates are in the lakes <30 km from Halifax in both directions. Other estimates of [ACID] SO_4 in lakes <30 km from Halifax made in a previous study (72±27 µeq/l) and that of the present study (65±21 µeq/l) are in reasonable agreement. However, both these estimates are substantially lower than those calculated from the data of Gorham (1957) and Watt et al. (1979), 80±27 and 95±21 µeq/l respectively (<40 km from Halifax).

In a previous study by Wiltshire and Machell (1981), lakes in the Halifax area compared with 11 other lakes outside the Halifax area in mainland Nova Scotia and southern New Brunswick, on the basis of Henriksen's empirical curve, have shown a greater acidification. On this basis, acidification is also clearly indicated in the present data with only two exceptions in a total of 36 lakes (Figure 2). Lakes in the present study, as a group, do not show a statistically significant higher acidification than other areas of the province based on Henriksen's empirical equations. However, Halifax lakes <30 km from the urban area do show a consistently higher level of acidification based on [ACID] SO_4 calculations, in this and our previous study.

3.4 The Effect of Distance from Halifax

The results of regressing pH, excess [SO_4], [Ca] + [Mg] and Gran alkalinity with distance from Halifax are given in Table 3 for the western lakes and in Table 4 for the eastern group. In the western group, slopes of the regression lines are significant for only alkalinity. A significant ($p = \leq 0.05$, $r = 0.51$) decline in alkalinity with increasing proximity to Halifax is suggested, although the regression coefficient indicates that a relatively small portion of the observed variation (26%) can be explained by the influence of distance. The regression of excess sulphate with distance is not significant ($p = \geq 0.05$, $r = 0.38$) in the lakes west of the city. There was no significant correlation between pH and distance, nor with the sum of [Ca] and [Mg]. The low correlation ($r = \leq 0.08$) of the latter parameters support the assumption that the influence of geology on water quality does not vary with distance throughout the western transect.

The eastern lakes show a significant relationship with distance for alkalinity ($p = \leq 0.05$) and for excess sulphate ($p = \leq 0.025$). Regression coefficients indicate that only 17.6% of the observed variation in Gran alkalinity is explained by distance, while 38.4% is explained by

TABLE 3 - RESULTS OF REGRESSING LAKE CONCENTRATIONS WITH LOG DISTANCE (km) WEST OF HALIFAX

Parameter	N	a	b	r	.025[1]	.050[2]
pH (-log [H^+])	18	4.312	0.455	0.35	NS	NS
Gran Alkalinity µeq/l	18	-49.127	27.920	0.51	NS	S
Excess Sulphate Log µeq/l	18	2.072	-0.204	0.38	NS	NS*
Excess [Ca] + [Mg] µeq/l	18	28.796	18.734	0.08	NS	NS

* Slope significant at p = $\leq$.10

TABLE 4 - RESULTS OF REGRESSING LAKE CONCENTRATIONS WITH LOG DISTANCE (km) EAST OF HALIFAX

Parameter	N	a	b	r	.025[1]	.050[2]
pH (-log [H^+])	18	4.083	0.448	0.35	NS	NS
Gran Alkalinity µeq/l	18	-50.328	22.315	0.42	NS	S
Excess Sulphate Log µeq/l	14	2.698	-0.616	0.62	S	S
Excess [Ca] + [Mg] µeq/l	17	43.698	0.670	0.01	NS	NS

[1] Slope different from zero at $\leq$0.025 level of significance.

[2] Slope different from zero at $\leq$0.05 level of significance.

distance for excess sulphate. Excess [Ca] + [Mg] show no relationship with distance and indicate a homogeneous effect of geology on water chemistry of lakes on the eastern transect. Thus, changes in geology do not appear to explain changes in alkalinity related to distance from Halifax. Distance also has no significant influence on the pH of the eastern lakes in the present data set. Furthermore, distance from Halifax has little influence on TOC levels in the eastern lakes (r^2 = 0.12), and on lake aluminum levels.

Although excess sulphate in the eastern lakes appears to decrease with increasing distance from Halifax, this relationship is heavily influenced by four lakes within 30 km of Halifax. If these lakes were not considered in the analysis there would be no significant relationship with distance (r^2 = .03) in the remaining lakes (>30-80 km). Watt (1979), reports a significant regression of excess sulfate on distance from Halfiax (r^2 = 0.54) although an inspection of the data suggests that the dependence on distance is created in the regression by lakes <30 km from Halifax and is also not apparent beyond 30 km.

The regression of distance vs excess sulfate for the eastern lakes predicts concentrations of 120 μeq/1 (5.7 ppm) at 10 km and 61 μeq/L (2.9 ppm) at 30 km from Halifax. The regression (non-log) given by Watt _et_ _al_. (1979) predicts excess [SO_4] of 125 μeq/1 (6 ppm) at 10 km which agrees well with the present estimate and reported lake concentrations. Watt's estimate of 88 μeq/1 (4.2 ppm) at 30 km is somewhat higher than that of the present study and recorded [SO_4] values. For all thirty-six lakes excess [SO_4] averaged 63 μeq/1 (3.0 ppm). Our estimate of 61 μeq/L (2.9 ppm) at 30 km supports the conclusion that little if any increase in excess [SO_4] occurs in the lakes beyond 30 km from Halifax.

Lake organic content appears to have little influence on excess sulphate levels. In all lakes >30 km from Halifax measurements of TOC and color do not correlate well with excess sulphate ($r^2 = 0.05$ and $r^2 = 0.12$ respectively). This is also the case in lakes close to Halifax (<30 km). Based on equations given in Oliver and Slawych (1981), the average concentration of organic anions has been estimated as 39 µm/l (48-30 µm/l) in the eastern lakes <30 km and 56 µm/l (17-76 µm/l) >30 km from Halifax. This confirms the fact that organic acids have a diminished influence on the water quality of the study lakes <30 km from the city and supports the conclusion that the chemical differences observed with increasing proximity to Halifax are related to anthropogenic influences and not natural factors.

The larger amounts of excess sulphate (approximately 59 µeq/l) close to Halifax cannot be accounted for by increased organic content ($[SO_4]$ from natural terrestrial sources) nor is this quantity completely balanced in our samples by increased levels of cations. Total cations (based on the mean of individual non-marine constituents) in lakes within 30 km of the city are however significantly higher than in lakes >30 km east of Halifax by 15-23 µeq/l. In the eastern lakes, a significant ($p = \leq 0.025$) relationship exists between higher excess sulphate levels and lower Gran alkalinities (alkalinity = $43.7914 - 32.2096 \log [SO_4]$, $r = 0.57$). From this it is estimated that 10-12 µeq/l of the increased excess sulphate in lakes <30 km east of the city is balanced by a corresponding increase in acidity (or negative Gran alkalinity). This increase in acidity corresponds to a pH decrease of approximately 0.22 units in lakes between 10 and 30 km from the city. The remaining increase in cations is created by slightly higher levels of excess calcium and aluminum. If the highly coloured lakes (>70 units) are removed from this analysis, increased acidity and aluminum account for this increase in cations. Within 30 km of Halifax, however, cations remain in deficit and in the present samples do not account for at least half of the additional 59 µeq/l of excess sulphate.

In the west, alkalinity decreases significantly with increasing proximity to Halifax, although absolute levels are higher than in the east. The predicted differences in negative Gran alkalinity (acidity) between 10 and 30 km is 9 µeq/l, similar to that in the east. However, corresponding excess sulfate levels are much lower, and no relationship with distance nor dependence ($p = \geq 0.05$) with alkalinity level is shown. A level of acidification similar to the eastern lakes is suggested by [ACID] SO_4 calculations. It is concluded that, although the effect of Halifax emissions is less significant in the west, there is evidence of acidification inside 30 km and there are indications that it is associated with a slight increase in excess [SO_4], most of which is balanced by the increased acidity. However, the large excess observed in SO_4 concentrations in the east is not seen.

4. CONCLUSIONS

The lakes in the study area are dilute, low in pH with a mean of 4.7 to 4.9 and with little, if any, buffering capacity (mean alkalinity of -7 to -15 µeq/l). These lakes are extremely sensitive to acid producing atmospheric inputs of anthropogenic origin, either local or long range. At present the effect of local sources of acidic or acid producing emissions on water quality appears to be restricted to the areas <30 km from the city and is significant east of Halifax. In the east the effects suggested in the present data are an increase of 59 μeq/l in excess [SO_4] (2.8 ppm) and an increase in acidity (negative Gran alkalinity) of 11 μeq/l.

This is a doubling of excess [SO_4] concentrations, which results in a small but significant increase in acidity and a small (0.2 units) but not significant decrease in lake pH with increasing proximity (<30 km) to Halifax.

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ACKNOWLEDGEMENTS

The authors gratefully acknowledge the assistance of D. Walter in the collection of samples. We also thank D. Walker and G. Platter of the Canadian Coast Guard for their provision of the helicopter which was instrumental in the collection of the samples. W. Horne, Dr. O. Vaidya and M. MacIssac are thanked for assistance in providing the chemical analyses. M. Keeling, B. Pellerin and P. Miller also deserve thanks for the typing of this report. Dr. R. Shaw and J. Kozak are thanked for their helpful review of the manuscript.

APPENDIX TABLE 1 - LOCATION AND GEOLOGY OF THE STUDY LAKES

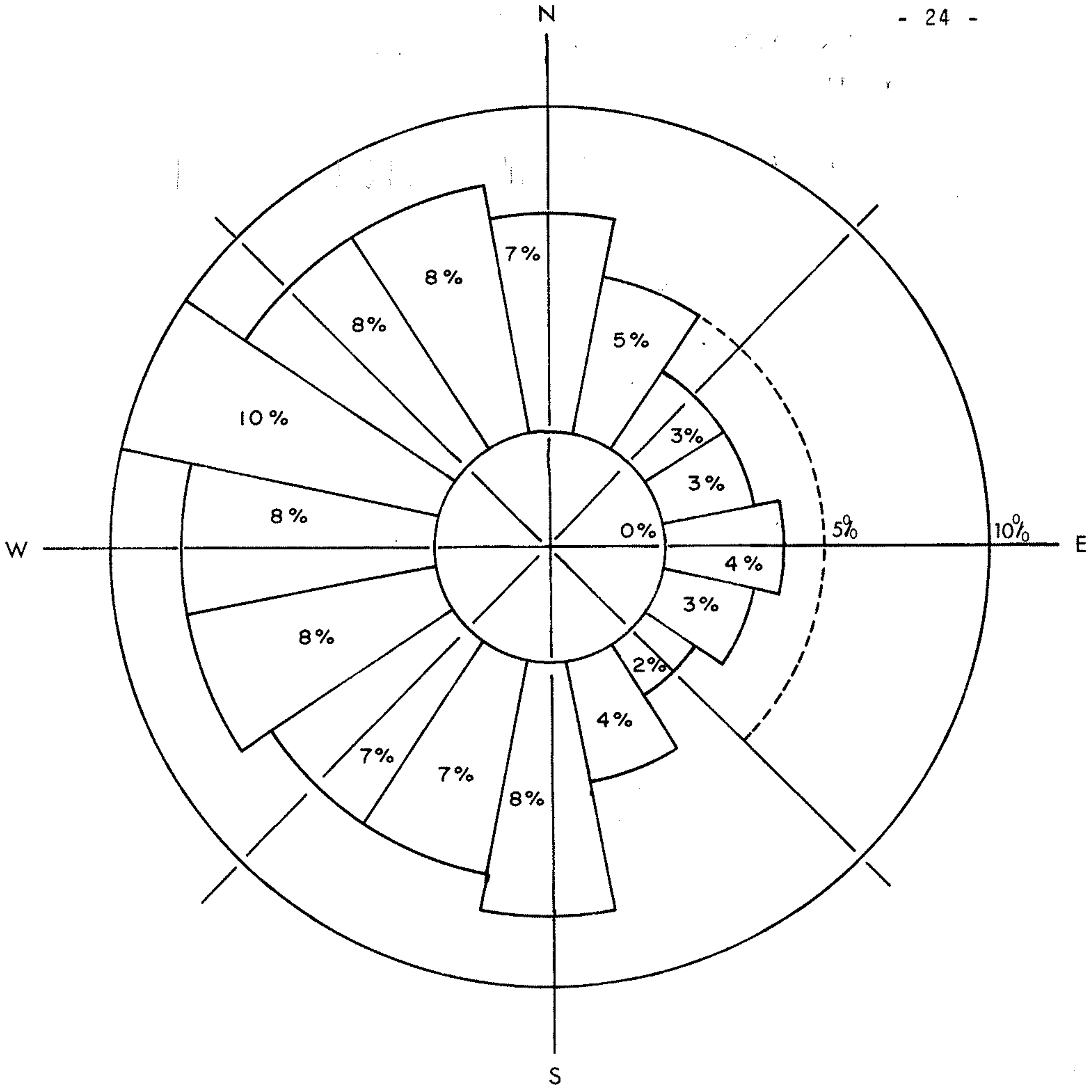
Lake No.	Lake Name	Coordinates Lat.	Coordinates Long.	km from Halifax	Bedrock Code
1	Ash Lake	44 40 30	63 14 15	10.4	Dgd
2	Otter Lake	44 37 30	63 43 30	11.9	DC mg
4	Sprucehill Lake	44 34 45	63 39 30	11	DC mg
5	Camphill Lake	44 41 45	63 49 00	19	Ca, DC mg
6	Hubleys Big Lake	44 38 40	63 50 00	20	DC mg
7	Big Five Bridges Lake	44 35 30	63 49 30	20	DC mg
8	Cooper Lake	44 44 30	63 51 20	22	Dgd
10	Big Black Lake	44 49 00	64 02 00	39	Dgd
11	Little St. Margarets Lake	44 45 50	64 06 00	42	DC mg
12	Armstrong Lake	44 49 30	64 11 30	51	DC mg
14	Chain Lake	44 50 00	64 19 30	61	EOg, Dgd
15	Card Lake	44 45 00	64 17 30	57	DC mg, Ca
16	Clean Waters Lake	44 49 00	64 22 50	65	EOg
17	Big Pine Lake	44 53 00	64 01 00	42	Dgd
20	Gold River Lake	44 51 45	64 29 00	74	Dgd
21	Big Connors Lake	44 43 20	63 57 45	31	DC mg
22	Green Lake	44 53 00	64 09 50	52	Dgd
23	Little Barrens Lake	44 47 25	64 08 35	46	DC mg
25/26 A	Duddley Lake	44 46 00	63 29 30	14	DC mg
25/26 B	East Lake	44 47 20	63 28 45	17	DC mg, EOg

APPENDIX TABLE 1 (Continued)

Lake No.	Lake Name	Coordinates Lat.	Coordinates Long.	km from Halifax	Bedrock Code
28	Preston Long lake	44 46 20	63 26 10	17	DC mg
30	Granite Lake	44 46 30	63 23 10	20	DC mg
32	Pace Lake	44 48 30	63 13 00	34	DC mg
33	Gibralter Lake	44 51 30	63 14 55	34	DC mg
34	Quaver Lake	44 50 00	63 14 15	34	DC mg
35	Spider Lake	44 51 30	63 10 20	39	DC mg
37	East Lake	44 52 45	63 10 45	40	DC mg
38	Little Duck Lake	44 48 30	63 01 45	47	DC mg
39	Bait Lake	44 49 00	62 57 20	53	DC mg
40	Blackley Lake	44 50 10	62 53 55	58	DC mg
42	Bear Lake	44 54 15	62 47 40	68	EOg, EOh
43	Paul Lake	44 51 45	62 48 10	66	DC mg
44	Chain Lake	44 50 50	62 45 35	69	DC mg
45	Second Lake	44 50 30	62 39 30	76	EOg
46	Henley Lake	44 51 55	62 39 30	77	DC mg
47	Davison Lake	44 51 45	62 44 00	71	DC mg

Bedrock Code:

Igneous Rocks:	DC - Devonian/Carboniferous
	D - Devonian
	g - Granite, mg - Monzo-Granite
	gd - Granodiorite
	Ca - Carboniferous
Metamorphic Rocks:	EOg - Goldenville Formation: Greywacke; Slate
	EOh - Halifax Formation: Slate; Siltstone



APPENDIX FIGURE 1

WIND ROSE SHEARWATER AIRPORT

Based on 18×R hourly averages: 1955 – 1972

APPENDIX TABLE 2 - ANALYTICAL DATA FOR THE HALIFAX AREA LAKES, JUNE 3, 1980 (Concentrations in mg/l)

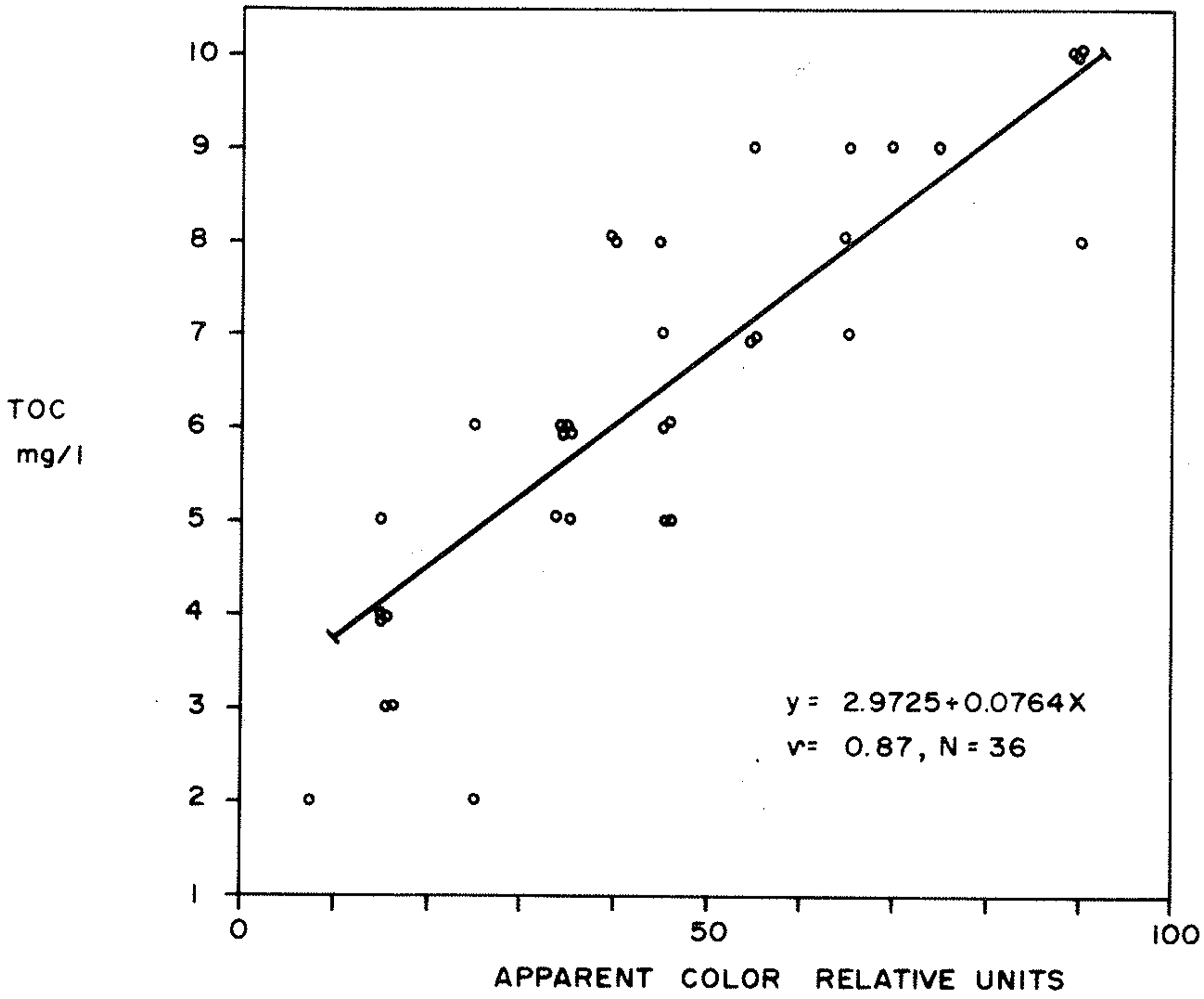
Lake	pH	Color Units	TOC	SO_4	Cl	Na/Cl	Na	Ca	Mg	K	Fe	Al	Mn	Cu	Zn	Pb	T °C	Sample
5	4.37	40-50	6.0	5.0	3.6	.72	2.6	0.44	0.39	0.27	.13	.33	.02	<.01	.01	<.02	16	070
28	4.38	50-60	7.0	5.0	5.7	.39	2.2	0.37	0.32	0.25	.15	.23	.01	<.01	<.01	.0	17	088
44	4.51	80-100	10.0	-	4.6	.65	3.0	0.30	0.38	0.31	.15	.38	.01	<.01	<.01	<.02	17	051
A	4.52	20-30	6.0	6.0	6.0	.40	2.4	0.61	0.41	0.27	.09	.25	.04	<.01	<.01	<.02	16.5	085
4	4.53	10-20	5.0	6.0	4.7	.64	3.0	0.47	0.49	0.27	.11	.28	.04	<.01	<.01	<.02	16	069
11	4.55	80-100	10.0	3.5	3.5	.69	2.4	0.59	0.29	0.25	.12	.23	.02	<.01	.01	<.02	17	075
47	4.56	80-100	10.0	<2.0	4.8	.60	2.9	0.29	0.30	0.27	.11	.27	.01	<.01	.01	<.02	16	054
37	4.57	80-100	8.0	4.0	4.0	.60	2.4	0.37	0.31	0.41	.22	.31	.02	.01	.03	<.02	16	058
B	4.58	10-20	4.0	6.0	5.3	.45	2.4	0.45	0.37	0.27	.04	.26	.02	<.01	<.01	.0	18	086
30	4.61	40-50	5.0	6.0	6.3	.51	3.2	0.62	0.39	0.31	.07	.23	.03	<.01	<.01	<.05	18	089
40	4.65	80-100	10.0	-	5.4	.57	3.1	0.38	0.38	0.31	.15	.34	.01	<.01	.01	<.02	-	058
7	4.65	60-70	8.0	3.0	5.0	.52	2.6	0.43	0.34	0.31	.12	.21	.01	.01	.01	.02	16	072
35	4.67	60-80	9.0	2.0	3.3	.82	2.7	0.58	0.37	0.35	.14	.23	.03	<.01	.01	<.02	16	063

APPENDIX TABLE 2 (Continued)

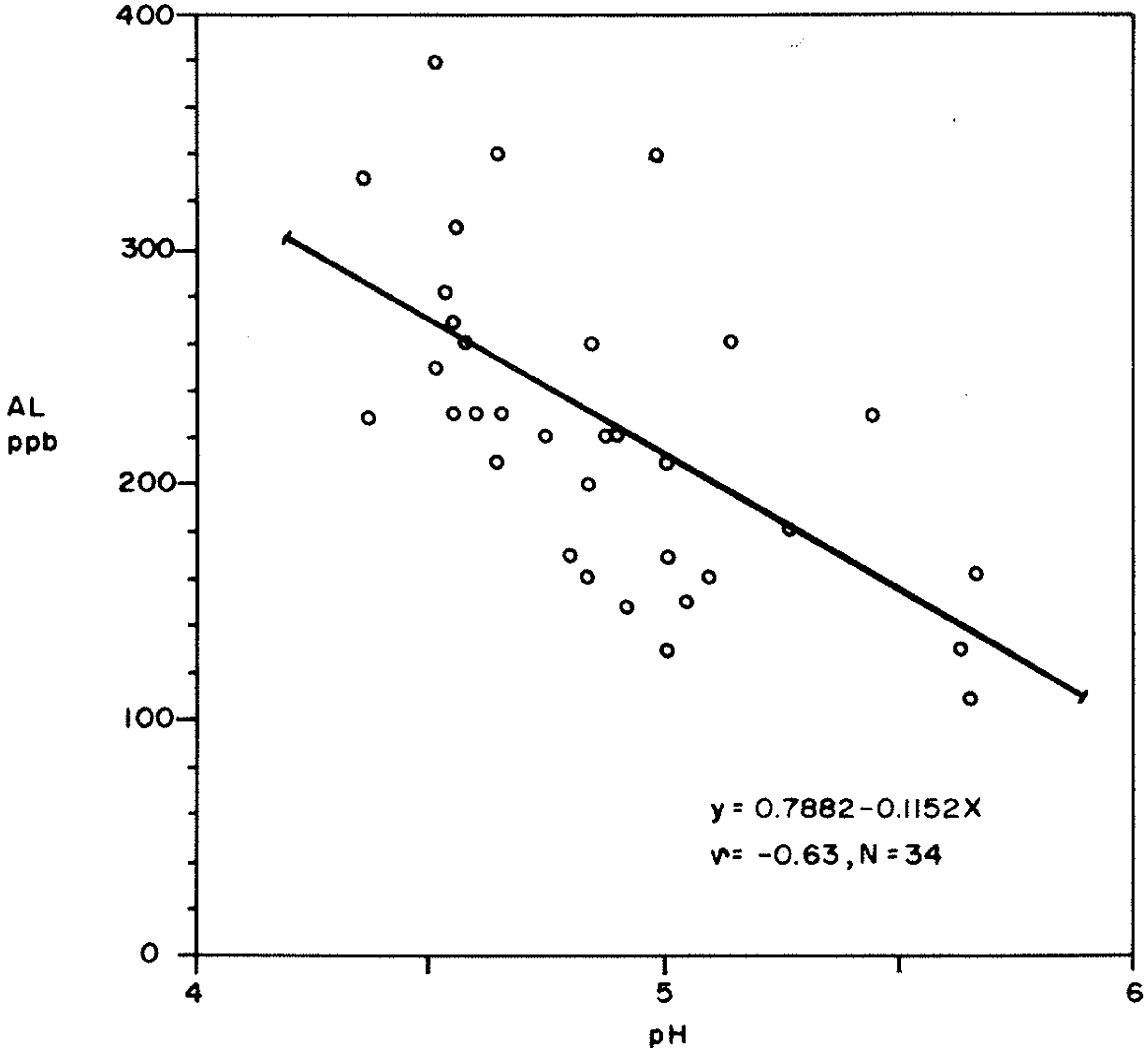
Lake	pH	Color Units	TOC	SO_4	Cl	Na/Cl	Na	Ca	Mg	K	Fe	Al	Mn	Cu	Zn	Pb	T °C	Sample
34	4.75	40-50	6.0	4.0	3.2	.94	3.0	0.48	0.36	0.41	.10	.22	.02	<.01	<.01	<.02	16	066
6	4.80	40-50	5.0	4.5	7.4	.61	4.5	0.75	0.41	0.35	.14	.17	.03	<.01	.01	<.02	17	071
23	4.84	40-50	8.0	4.0	3.5	.57	2.0	0.40	0.25	0.27	.07	.16	.01	<.01	<.01	<.02	17	084
16	4.85	10-20	4.0	2.0	2.9	.83	2.4	1.33	0.54	.25	.03	.04	.01	<.01	<.01	.02	16.5	079
45	4.85	30-40	6.0	4.0	5.6	.57	3.2	0.44	0.46	0.27	.11	.20	.02	<.01	.01	<.02	17	052
42	4.85	70-80	9.0	-	4.0	.60	2.4	0.51	0.31	0.27	.20	.26	.03	<.01	.01	<.02	16	055
21	4.89	30-40	6.0	3.5	7.4	.39	2.9	0.45	0.41	0.75	.09	.22	.03	<.01	<.01	<.02	17	082
43	4.90	50-60	7.0	4.0	4.2	.76	3.2	0.50	0.40	0.35	.17	.22	.05	<.01	<.01	<.02	16	060
46	4.92	20-30	2.0	-	4.9	.57	2.8	0.35	0.34	0.27	.04	.15	.06	.01	.02	<.02	16	057
15	4.99	30-40	6.0	3.5	4.0	.73	2.9	0.63	0.36	0.41	.26	.34	.05	<.01	.01	<.01	16	078
1	5.01	5-10	2.0	3.7	2.8	.93	2.6	0.54	0.36	0.41	.01	.17	.04	<.01	.01	<.02	16	067
2	5.02	10-20	3.0	3.0	4.0	.75	3.0	0.57	0.44	0.25	.22	.13	.10	<.01	<.01	<.02	16	068
12	5.03	60-70	7.0	3.0	3.1	.74	2.3	0.72	0.31	0.27	.19	.21	.02	<.01	<.01	<.02	16.5	076

APPENDIX TABLE 2 (Continued)

Lake	pH	Color Units	TOC	SO_4	Cl	Na/Cl	Na	Ca	Mg	K	Fe	Al	Mn	Cu	Zn	Pb	T °C	Sample
17	5.06	40-50	8.0	4.0	4.6	.57	2.6	0.97	0.36	<.25	.09	.15	.04	<.01	<.01	<.02	17	080
33	5.10	30-40	5.0	<2.0	4.6	.67	3.1	0.67	0.39	0.41	.05	.16	.02	<.01	.01	<.02	15.5	062
38	5.14	30-40	6.0	3.5	4.4	.75	3.3	0.68	0.46	0.41	.08	.26	.04	<.01	<.01	<.02	16.5	064
32	5.16	10-20	3.0	3.5	4.0	.80	3.2	-	0.1	0.41	-	-	<.01	<.01	<.01	<.02	14	065
20	5.20	50-60	9.0	4.0	5.9	.44	2.6	0.99	0.46	0.27	.12	.17	.02	<.01	.01	<.02	17	081
10	5.28	30-40	5.0	3.5	2.9	.72	2.1	0.74	0.31	<.25	.05	.18	.03	<.01	<.01	<.02	16	074
14	5.44	60-70	9.0	3.5	3.0	.77	2.3	1.13	0.47	0.27	.15	.23	.03	<.01	<.01	<.02	17	077
8	5.63	10-20	4.0	7.5	-	.32	5.6	1.60	0.59	0.6	.02	.13	.07	<.01	<.01	<.02	16	073
22	5.66	10-20	4.0	3.5	3.8	.58	2.2	0.79	0.30	<.25	.03	.11	.01	<.01	<.01	<.02	17	083
39	5.67	40-50	7.0	<2.0	4.6	.78	3.6	1.22	0.52	0.41	.04	.16	.03	<.01	<.01	<.02	16	061



APPENDIX FIGURE 2
THE RELATIONSHIP BETWEEN COLOR AND TOC FOR
THE STUDY LAKES



APPENDIX FIGURE 3
THE RELATIONSHIP BETWEEN pH AND TOTAL ALUMINUM
IN THE STUDY LAKES

APPENDIX TABLE 3 - ESTIMATES OF ACIDIFICATION IN THE INDIVIDUAL STUDY LAKES BASED ON HENRIKSEN'S EMPIRICAL RELATIONSHIPS

	Lake	D (km)	Sample No.	pH	(ALK) μeq/l	xs (Ca) μeq/l	xs (SO_4) μeq/l	(ACID) Ca μeq/l	(ACID) SO_4 μeq/l	Color	TOC mg/l
1	Ash	10.4 W	067	5.01	-5	24	69	8	49	5-10	2
4	Spruce Hill	10.5 W	069	4.53	-34	19	111	29	87	10-20	5
2	Otter	11.9 W	068	5.02	-7	24	51	10	33	10-20	3
25	Dudley (A)	13.5 E	085	4.52	-24	24	108	27	84	20-30	6
26	East (B)	16.5 E	086	4.58	-24	17	110	17	86	10-20	4
28	Preston Long	16.8 E	088	4.38	-44	12	88	31	66	50-60	7
5	Camphill	18.5 W	070	4.37	-42	18	94	37	71	40-50	6
6	Hubley's Big	19.6 W	071	4.80	-14	30	72	24	52	40-50	5
30	Granite	20.2 E	089	4.61	-24	24	107	27	83	40-50	5
7	Big Five Bridges	20.3 W	072	4.65	-19	16	48	11	31	60-70	8
8	Cooper	22.9 W	073	5.63	4	61	105	48	81	10-20	4
21	Big Connors	30.5 W	082	4.89	-4	15	51	-6	34	30-40	6
34	Quaver	33.5 E	066	4.75	-18	21	74	16	54	40-50	6

APPENDIX TABLE 3 (Continued)

Lake	D (km)	Sample No.	pH	(ALK) μeq/l	xs (Ca) μeq/l	xs (SO_4) μeq/l	(ACID) Ca μeq/l	(ACID) SO_4 μeq/l	Color	TOC mg/l
32 Pace	34.3 E	065	5.21 5.16	2 4	<-4	61	<-36	43	10-20	3
33 Gibraltar	34.4 E	062	5.10	0	29	<28	9	<13	30-40	5
35 Spider	39.4 E	063	4.67	-17	26	32	22	17	60-80	9
10 Big Black	39.4 W	074	5.28	-1	34	64	17	45	30-40	5
37 East	40.2 E	058	4.57	-26	14	72	16	52	80-100	8
11 Little S.M.B.	42 W	075	4.55	-22	26	63	27	44	80-100	10
17 Big Pine	42 W	080	5.06	-3	44	70	32	50	40-50	8
23 Little Barrens	46.3 W	084	4.84	-8	16	73	1	53	40-50	8
38 Little Duck	46.8 E	064	5.14	8	29	60	2	41	30-40	6
12 Armstrong	51 W	076	5.03	7	33	33	7	17	60-70	7
22 Green	51.5 W	083	5.66	9	35	62	9	43	10-20	4
39 Bait	53 E	061	5.67	8	56	<28	37	<13	40-50	7
15 Card	56.8 W	078	4.99	-2	27	61	9	43	30-40	6

APPENDIX TABLE 3 (Continued)

	Lake	D (km)	Sample No.	pH	(ALK) μeq/l	xs (Ca) μeq/l	xs (SO_4) μeq/l	(ACID) Ca μeq/l	(ACID) SO_4 μeq/l	Color	TOC mg/l
40	Blackley	57.8 E	053	4.65	-11	13	355	0	304	80-100	10
14	Chain	61.4 W	077	5.44	17	53	64	24	45	60-70	9
16	Cleanwaters	65.0 W	079	4.85	-12	63	33	67	18	10-20	4
43	Paul	65.8 E	060	4.90	-9	21	71	7	51	50-60	7
42	Bear	68.3 E	055	4.85	-11	21	320	10	272	70-80	9
44	Chain	68.8 E	051	4.51	-33	10	-	17	-	80-100	10
47	Davison	71.0 E	054	4.56	-19	9	< 28	2	< 13	80-100	10
20	Gold River	74.3 W	081	5.20	5	43	66	23	47	50-60	9
45	Second	76 E	052	4.85	-9	16	67	1	48	30-40	6
46	Henley	76.8 E	057	4.92	-10	12	159	-3	129	20-30	2