Fisheries and Oceans
Science

Pêches et Océans
Sciences

C S A S

Canadian Stock Assessment Secretariat

S C É S

Secrétariat canadien pour l'évaluation des stocks

Research Document 2000/055

Document de recherche 2000/055

Not to be cited without
permission of the authors [1]

Ne pas citer sans
autorisation des auteurs [1]

Freshwater chemistry acidification trends in sensitive Nova Scotia lakes: 1983-1997

Thomas A. Clair
Environmental Conservation Branch, Environment Canada
Atlantic Region, P.O. Box 6227
Sackville, N.B. E4L 1G8

James M. Ehrman
Digital Microscopy Facility, Mount Allison University
Sackville, N.B. E4L 1G7

and

Chul-Un Ro
Atmospheric Environment Service, Environment Canada
4905 Dufferin St.
Downsview, Ont. M3H 5T4

[1] This series documents the scientific basis for the evaluation of fisheries resources in Canada. As such, it addresses the issues of the day in the time frames required and the documents it contains are not intended as definitive statements on the subjects addressed but rather as progress reports on ongoing investigations.

Research documents are produced in the official language in which they are provided to the Secretariat.

This document is available on the Internet at:

[1] La présente série documente les bases scientifiques des évaluations des ressources halieutiques du Canada. Elle traite des problèmes courants selon les échéanciers dictés. Les documents qu'elle contient ne doivent pas être considérés comme des énoncés définitifs sur les sujets traités, mais plutôt comme des rapports d'étape sur les études en cours.

Les documents de recherche sont publiés dans la langue officielle utilisée dans le manuscrit envoyé au Secrétariat.

Ce document est disponible sur l'Internet à:

http://www.dfo-mpo.gc.ca/csas/

ISSN 1480-4883
Ottawa, 2000

Abstract

Nova Scotia lakes have been sampled at least twice yearly since the spring of 1983 for a number of water chemistry parameters. Statistical analysis of a number of the water chemistry variables was done in 1998 to see if trends could be detected in the main ions which caused acid precipitation or which could be dependent on changes in acidification. The data show consistent results. We could detect no long-term trends in pH, acid neutralization capacity (ANC) or dissolved organic carbon. We show a decrease in long-term sulfate and in total sum of cations. The latter two parameter trends suggest two things: a) that the main acidifying ion seems to be decreasing in the system, and b) that the weathering caused by the sulfuric acid is also decreasing. Our results thus suggest that long-term chronic acidification is decreasing, but not enough to change pH or ANC. We also compare the results of this trend analysis to previous ones which were done on 1983-91 and 1983-1994 data series.

Résumé

Depuis le printemps 1983 et au moins deux fois l’an, on a prélevé des échantillons dans des lacs situés en Nouvelle-Écosse afin de déterminer un certain nombre de caractéristiques chimiques de l’eau. En 1998, on a effectué l’analyse statistique d’un certain nombre de variables relatives à la chimie de l’eau afin de déterminer si on pouvait établir des tendances relatives aux ions principaux qui causent les précipitations acides ou qui pourraient dépendre des variations de l’acidification. Les données produisent des résultats compatibles. Nous n’avons pas pu établir une tendance à long terme en ce qui concerne le pH, le potentiel de neutralisation de l’acide (PNA) ou la teneur en carbone organique dissous. Nous avons observé une diminution à long terme de la teneur en sulfate et de la concentration totale de cations. Les tendances relatives à ces deux paramètres suggèrent deux hypothèses: a) que la concentration du principal ion acidifiant dans le système semble diminuer, et b) que l’altération atmosphérique causée par l’acide sulfurique diminue elle aussi. Les résultats que nous avons obtenus suggèrent donc que l’acidification chronique et à long terme diminue, mais pas assez pour avoir un effet sur le pH ou le PNA. Nous comparons aussi les résultats de cette analyse de tendances à ceux d’analyses antérieures qui ont été réalisées à l’aide des séries de données des périodes 1983-1991 et 1983-1994.

Introduction

Acid precipitation has been a serious environmental problem in eastern North America and in Atlantic Canada in particular. Research has shown that Atlantic salmon (*Salmo salar*) populations have been reduced in the latter part of the century in this region and that the reduction is most likely due to increases in the deposition of atmospheric sulfate in the region [Watt, 1987]. More recent work suggests that salmon populations, especially in the Eastern Shore of Nova Scotia, have continued this precipitous decline in the last decade.

Nova Scotia has been intensively studied for acidification impacts. The region is especially sensitive to the problem, as it receives acid precipitation originating from all the major production regions of North America [Shaw, 1979]. Much of the area is also underlain by slates and other bedrock types which weather poorly and thus have poor acid buffering capacities. Atlantic Canada surface water pH values reach some of the lowest measured in North America [Kerekes *et al*., 1982].

In 1983, a lake monitoring network was established in the Yarmouth and Kejimkujik regions of Nova Scotia in order to allow the analysis of long-term trends in acidification effects on freshwaters. Analysis of acidification trends was done on these lakes on two previous occasions [Clair *et al.*, 1995; Jeffries, 1997]. In our first analysis we showed that precipitation sulfate (SO_4^{-2}) and hydrogen ion (H^+) concentrations decreased from 1983 to 1991 in Nova Scotia. We found that though surface water SO_4^{-2} increased, pH and acid neutralization capacity (ANC) also increased. The second analysis was done on 1983-94 data [Jeffries, 1997] and showed a change in trends as some reductions in water SO_4^{-2} were noted, with little change in other variables. Because of these conflicting results, it was therefore unclear whether there were actual long-term improvements in the acid-base status of the lakes.

In order to cover gaps in our spatial coverage, we also began a sampling program in lakes from the Eastern Shore and Cape Breton regions in 1989 and report results of trend analysis for 1989-1997 in order to see if there were changes occurring in this critical region over the last decade. This report provides results of trend analysis for these regions for the first time.

We tested our data to see if there were significant trends in acidification variables (pH, sulfate, nitrate, ANC and base cations) over the whole 1983 to 1997 and from 1989 to 1997 in the lakes from Kejimkujik and Yarmouth areas, as well as from 1989 to 1997 in the Eastern Shore region. Though these data are from lakes and not rivers, they nevertheless indicate trends in regional geochemical processes which are also operating with the rivers. The chemical interactions demonstrated in these lake waters are also the same as those which affect the rivers, and thus can be used as indicators of regional processes.

Area and Methods

Much of Nova Scotia is underlain by poorly weatherable bedrock such as granites and slates with little acid buffering capacity. The area was glaciated during the last ice age (approx. 10,000 years BP) and thus soils overlying the bedrock are thin and acidic in much of the region [Ehrman *et al*., 1993]. Initially, 40 lakes located in Nova Scotia were identified as relatively free of anthropogenic contact and were deemed "typical" of their region [Clair and Esterby, 1991]. Lakes were not selected randomly over the region using a grid approach, but were chosen in regions where large numbers of sites were clumped as this provided some efficiencies in sampling costs. We thus sampled lakes in the regions around Kejimkujik National Park in southwestern Nova Scotia (Fig. 1). The initial lakes were felt not to fully represent the Nova Scotia region geographically, so that a further 31 lakes were added in 1989, located east of Halifax in the central part of the Province, and also in the Cape Breton Highlands National Park, at the far eastern end of the region (Fig. 1).

All lakes were sampled twice yearly, in the spring and fall water overturn periods. This allowed us to collect well mixed waters without the complications introduced by lake thermal stratification. Samples were usually collected using a helicopter. At every 10th site sampled, triplicates were collected and compared to each other to ensure that no chronic contamination or laboratory problems existed. We used only the median values of the triplicates in our analysis. The samples were analyzed at the Environment Canada Laboratory in Moncton, New Brunswick, for the following chemical parameters: specific conductance, pH (potentiometric), acid neutralization capacity (ANC, using the Gran method), sulfate (SO_4^{-2}), chloride, nitrate (NO_3^-) (ion chromatography), calcium, magnesium (atomic absorption), sodium, and potassium (flame emission). This laboratory was part of Environment Canada’s Laboratory Quality Assurance Program and was usually rated as one of the better labs in the country for acid rain - related samples.

We also analyzed dissolved organic carbon (DOC), a surrogate for organic acidity, using the persulfate-UV oxidation method from 1983 to 1993 and the high temperature combustion method from 1994 to 1997. The two methods did not produce comparable results as the earlier method tended to underestimate DOC by approximately 25% [Koprivnjak *et al*., 1995].

A precipitation sampling station in Environment Canada's, Atmospheric Environment Service CAPMoN network, was located in Kejimkujik in Nova Scotia and Baie D'Espoir in Newfoundland (Fig. 1). Samples were collected and analyzed using standard AES protocols. Wet deposition data was summarized to produce annual volume-weighed concentrations and deposition calculations [Ro *et al*., 1998].

As most of these sites are within 100 km of the Atlantic Ocean, seaspray can be an important component of the precipitation and lake water chemistry. Chloride (Cl^-) in water can be used as a tracer of the seaspray effect as it is not a common constituent of bedrock minerals or soils. Assuming that all Cl^- measured is of marine origin, and assuming that the standard seawater $Cl^-:SO_4^{-2}$: $Na^+:Ca^{+2}:K^+:Mg^{+2}$ ratios apply [Watt *et al*., 1979], we use them to identify the fraction of these ions which are from seaspray and remove them from the total value. This gives an accurate picture of ion movement caused by acidification or normal weathering. As the cations released are dependent on the type of mineral being weathered and as we generally don't know what that is, we sum all sea-salt corrected base cations into one variable, base cations (C_b), reported in $\mu eq\ L^{-1}$ to get an overall value of weathering impact. We did trend analysis only on seasalt corrected values.

A number of methods have been used in the past to analyze water quality trends. However, due to problems associated with data autocorrelation and seasonal cycles, the analysis of trends in water chemistry data sets has been shown to be best done with non-parametric methods [Berryman *et al*., 1988; Hirsch and Slack, 1982; Clair *et al*., 1995]. The methods may not be as statistically powerful at Box-Jenkins analysis [Clair and Whitfield, 1983], or as some linear regression methods e.g. Sirois [1993] but have been found to be robust and requiring few assumptions regarding data distribution.

We analyzed data using two time windows. First, we calculated trend statistics for 1983 to 1997 data sets from the 36 Nova Scotia (NS) for which we had complete series. We then analyzed the data from the newer 31 NS lakes which were sampled from 1989 to 1997 and reanalyzed the data from the 36 NS sites using only the 1989 to 1997 portions of the data. We also analyzed the annual ion deposition results (CAPMoN data) for trends using the same statistical methods described above.

In order to determine if there were regional tendencies for each variable, we compared the numbers of increasing versus decreasing trends for each lake. If, for example, more than twice as many statistically significant trends showed an increase as opposed to a decrease over a certain period of time, we then assumed that the overall regional direction was increasing. If the number of increasing and decreasing trends were roughly equal, we assumed no regional trend. We also obviously assumed that sites which showed no trend direction did not disprove the null hypothesis of no change.

Results

Atmospheric deposition

Using our non-parametric approach, SO_4^{-2} deposition data showed a measurable decrease from 1983 to 1997 in Nova Scotia, from 324 to 214 eq ha^{-1} in NS (Table 1). There was no statistical SO_4^{-2} trend at Kejimkujik over the 1989-97 period, even though a regression line is visually obviously steeper than the long-term line. The lack of a trend is due to the greater scatter in a shorter data set. With some long-term change in SO_4^{-2}, we therefore expected some decreases in H^+ deposition. This partially happened, as the non-parametric approach showed Kejimkujik deposition to decrease significantly only from 1989 to 1997 (Fig. 2). Deposition average for the first three years of the record (1984-1986) at Kejimkujik were 440 eq ha^{-1} and for the last three years (1995-1997), 310 eq ha^{-1}.

No trends could be detected in NO_3^- deposition at the monitoring sites, which seems to match the fact that North American production has not changed very much [Brook, 1997]. In the first three years, Kejimkujik NO_3^- deposition averaged 169.5 eq ha^{-1} versus 157.12 eq ha^{-1} at the end. There also was no trend in C_b deposition at either site.

Lake chemistry trends

Over the period when the monitoring program has been operating (1983-97), lake H^+ concentrations did not reflect the reduced sulfate deposition (Table 2). Sites showed little in the way of H^+ trends over this time, as one site showed increased concentrations, one decreased, and the remaining 34 showed no change. In the 1989-97 time window, NS lakes showed a major change in trend directions. Nine sites showed an H^+ increase, while only two showed decreases. The lake increases were distributed throughout NS and were not confined to just one area (Fig. 3).

Lake SO_4^{-2} trends, showed a general decrease (Table 2), through both the long and short-term time windows (e.g., Fig. 4). Sixteen of 36 (44%) lakes showed decreasing values over the whole series. In contrast, 56 of 67 lakes (84%) showed decreasing SO_4^{-2} trends in the 1989-97 portion of the Nova Scotia record. Again, these were well distributed throughout the Province (Fig. 3). Lake SO_4^{-2} thus showed the pattern we expected with the decrease in deposition, though the regional trend was stronger over the last few years of the record in NS.

The lakes showed some increase in ANC over the 1983-97 period, but this was a pattern which was reversed over 1989-97, when six lakes showed decreases and only three had increases, corroborating the H^+ change, but contradicting the SO_4^{-2} decrease.

Base cation concentrations showed little change over the length of the 1983-97 record. Examination of 1989-97 series, however, indicates that 15 lakes (22%) showed decreasing concentrations, in line with decreased weathering (e.g., Fig. 4). Two lakes showed increases and 48 had no change. As with SO_4^{-2} and H^+, C_b shows a large change in trends in the latter part of the record.

Discussion

Trends

Our data clearly show the importance of a phenomenon which was explored by Ehrman and Clair [1995]. They showed that the data window used to study trends often made a significant difference to whether or not trends could be detected and could also determine the direction of that trend. In other words, using longer data sets will often obscure shorter trends occurring within the series. It is therefore important to clearly define the objectives of the analysis in order to better interpret the results. In our case, as we were interested in determining if emission controls were making an impact on water chemistry, the shorter series were clearly more useful in seeing how basin geochemistry was behaving. The longer time series, on the other hand, incorporated a great deal of geochemistry which occurred before the period where enhanced emission controls were instituted.

Despite decreasing atmospheric SO_4^{-2}, H^+ is increasing in a number of lakes in the latter portion of the NS record (Fig. 3). This is in contrast to the few decreasing trends we measured with the 1983-91 record, when H^+ decreased somewhat in Nova Scotia. Sulfate decreased over both the 1983-97 and 1989-97 series in NS (e.g., Fig. 4), though our analysis shows a very strong trend toward lower SO_4^{-2} with the later time window. This contrasts with the 1983-91 data which showed increases. These results confirm a tendency of decreasing SO_4^{-2} in the region which began with the 1983-94 data.

C_b trends in NS show a decrease over the last portion (Fig. 4) of the record in NS and no trend from 1983 to 1997. Overall then, there seems to be a short-term trend in base cation decrease in NS.

Finally, ANC showed few consistent patterns in both recent short and long-term data. There was a strong increase in ANC in the earliest sampling period (1983-91), though this either was either dampened or reversed over the longer term. In our data set, ANC trends are inconsistent and seem to be easily changed from one direction to another.

Overall, an analysis of time window results for all variables allows us to see that there can be short-term trends within long-term series. H^+, SO_4^{-2}, ANC and C_b all show differences in trend directions between the earliest analyzed series (1983-91) and the latest series (1989-97). Therefore, it is clear that we can lose a great deal of information if we analyze long series without considering short-term changes.

Geochemistry

Equation 1 described the general relationship between the ionic constituents in natural waters [Stumm and Morgan, 1996]. We should then expect the changes in trends measured in the different variables to conform to the electroneutrality law:

$$\varphi C_b - \varphi C_a = [ANC] = [HCO_3^-] + 2[CO_3^{-2}] + [Or^-] + [OH^-] - [H^+] \text{ (Eq. 1)},$$

where φC_b (sum of base cations) includes Ca, Mg, K, Na, φC_a (sum of acid anions) includes SO_4^{-2}, NO_3^-, Cl^-. HCO_3^- is bicarbonate, CO_3^{-2} is carbonate, OH^- is the hydroxyl ion and Or^- is the organic anion component, which is important in waters with DOC values > 5 mg l^{-1}.

Deposited H^+ from atmospheric H_2SO_4 weathers minerals in bedrock and soils to release calcium, magnesium, potassium or sodium, e.g.,

$$\underset{\text{(calcium feldspar)}}{CaAl_2Si_2O_8(s)} + 2H^+ + H_2O + SO_4^{-2} = Ca^{+2} + \underset{\text{(kaolinite)}}{Al_2Si_2O_5(OH)_4(s)} + SO_4^{-2} \text{ (Eq. 2)}$$

[Schnoor and Stumm, 1986].

As atmospheric H_2SO_4 decreases, two things should happen. First, SO_4^{-2} decreases in waters draining the lake basins. Secondly, as there should be less weathering occurring, fewer base cations such as Ca^{+2} (e.g., Eq. 2) will be released from the soils and bedrock and dissolved into streams and lakes. In our region, SO_4^{-2} and C_b decrease as theory would suggest. pH and ANC will therefore not begin improving in these basins until weathering from mineral acids associated with SO_4^{-2} is lower than that from the natural weathering [Reuss *et al.*, 1987].

Our results do not completely follow the expected pattern however, as H^+ increased and ANC decreased in Nova Scotia over the 1991-97 period, when they should have remained stable or have gone in opposite directions. The other main H^+ source in these waters is organic acidity which suggested to us that DOC in lake waters may have risen over the years, causing an increase in natural acidity. Unfortunately, the DOC data could not be used in the trend analysis to confirm this hypothesis due to a change in analytical methods which occurred in 1993 (see Methods). Clair and Freedman [1986] have shown that in this region, DOC concentrations are inversely correlated to precipitation, due to a concentration-dilution relationship which is common in these temperate systems. We therefore studied precipitation trends in our two regions, to see if they had decreased in the latter part of the record. This at least would suggest that changing climate variables might have an influence on organic acidity. Analysis of precipitation trends at the CAPMoN site over the 1983-97 and 1991-97 periods showed no significant trends however (Fig. 5), so that changes in organic acidity cannot be inferred from the available data.

The role of seasalt ion exchange also has not been understood yet in these waters. Norton and Kahl [1999] have shown that marine Na^+ exchanges with soil H^+, releasing it to streams. Our monitoring data is not suitable for evaluating this potential contribution however. More work will be done on calibrated basin data which is collected more frequently to better understand if this phenomenon is of importance in our case.

Our results partly match those of Clow and Mast [1999] who reported on data from five streams of the USGS Hydrologic Benchmark Network in the northeastern United States. Their study showed generally decreasing trends in acid deposition. Like us, they found that SO_4^{-2} and C_b tended to decrease in the streams. Unlike us, they found pH and ANC relatively unchanging. Stoddard *et al*. [1999] found that generally, SO_4^{-2} and C_b decreased in most regions of North America. However, they also found significant ANC decreases in the New York State and in the US Midwest. Our results are therefore not unique, even though they are somewhat counterintuitive.

The results from widely separated sites receiving varying amounts of SO_4^{-2} suggest that the path to ecosystem recovery from acidification is far from being simple or direct. Our atmospheric trend results show that SO_4^{-2} decreased mostly at the beginning of the record, while lake SO_4^{-2} and its companion weathering products decreased near the end. Our results confirm that because of drainage basin retention of major weathering byproducts and tracers, further reductions in the atmospheric deposition of SO_4^{-2} will most likely not be immediately reflected in pH and ANC, but will take some time to be reflected in basin geochemical outputs. We've also realized that measured increases in acidity and decreases in ANC are difficult to reconcile with the decrease in SO_4^{-2}.

This could be caused by changing climatic conditions which would affect organic acid concentrations or also due to marine salt influences. A better DOC data set, a surrogate for organic acidity, is currently being collected, and should help us resolve part of this problem in the future. Data collected weekly at calibrated basins in NS will help us address the marine salt issue.

These results clearly show that despite the introduction of significant control measures, acid deposition is still a problem for Nova Scotia's waters. The decrease of SO_4^{-2} and C_b in regional waters tells us that the emission reductions are having some impacts. However, pH and ANC which are key to aquatic life are not improving. This leads to the conclusion that critical loads in this region are still being exceeded and that more emission cuts are probably needed for a geochemical equilibrium to reestablish itself within a reasonable time span.

Acknowledgments

We thank the Warden staff at Cape Breton National Park for sampling lakes for us. The authors also thank the staff of the Environment Canada Laboratory in Moncton, New Brunswick for their high quality analytical work.

References

Berryman, D., B. Bobee, D. Cluis, and J. Haemmerli. 1988. Non-parametric tests for trend detection in water time series, Water Resour. Bull., 24, 545-556.

Brook, J. R. 1997. New scientific results, 1997 Canadian acid rain assessment, Vol. 2, Atmospheric Science Assessment report, Environment Canada, Downsview, Ont.

Clair, T. A. and B. Freedman. 1986. Patterns and importance of dissolved organic carbon in four acidic brownwater streams in Nova Scotia, Canada, Water, Air, Soil Pollut., 31, 139-147.

Clair, T. A., P. J. Dillon, J. Ion, D. S. Jeffries, M. Papineau, and R. J. Vet. 1995. Regional precipitation and surface water chemistry trends in southeastern Canada (1983-91), Can. J. Fish. Aquat. Sci., 52, 197-212.

Clair, T. A.and S. R. Esterby. 1991. Chemical grouping of Environment Canada's lake acidification monitoring sites in Atlantic Canada, Tech. Rep. IWD-AR-WQB-91-168, Environment Canada - Atlantic Region, Moncton, N.B., Canada.

Clair, T. A. and P. H. Whitfield. 1983. Trends in pH, calcium, and sulfate in Atlantic Canadian rivers, Limnol. Oceanogr., 28, 160-165.

Clow, D. W. and M. A. Mast. 1999. Long-term trends in stream water and precipitation chemistry at five headwater basins in the northeastern United States, Water Resour. Res., 35, 541-554.

Ehrman, J. M. and T. A. Clair. 1995. Step-wise analysis of precipitation and river chemistry trends in Atlantic Canada, Water, Air, Soil Pollut., 85, 671-676.

Ehrman, J. M., G. D. Howell, S. Roussel, and T. A. Clair. 1993. Summary of biophysical and water chemistry for 100 lakes in the Environment Canada Atlantic Region acid deposition network, Tech. Rep. WRD-AR-MEB-93-180, Environment Canada, Atlantic Region, Sackville, NB.

Hirsch, R. M. and J. R. Slack. 1982. Techniques of trend analysis for monthly water quality data, Water Resour. Res., 18, 107-121.

Jeffries, D. S. 1997. The Effects on Canada's lakes, rivers and wetlands, Vol. 3, 1997 Canadian acid rain assessment, Environment Canada, Burlington, Ont.

Kerekes, J., G. Howell, S. Beauchamps, and T. L. Pollock. 1982. Characterization of three lake basins sensitive to acid precipitation in Central Nova Scotia, Int. Rev. ges. Hydrobiol., 67, 679-694.

Koprivnjak, J. F., J. G. Blanchette, R. A. Bourbonniere, T. A. Clair, A. Heyes, K. R. Lum, R. McRae, and T. R. Moore. 1995. The underestimation of concentrations of dissolved organic carbon in freshwaters, Wat. Res., 29, 91-94.

Norton, S. A. and J. S. Kahl. 1999. Impacts of marine aerosols on surface water chemistry at Bear Brook watershed, Maine, Verh. Internat. Verein. Limnol.

Reuss, J. O., B. J. Cosby, and R. F. Wright, 1987. Chemical processes governing soil and water acidification (Review Article), Nature, 329, 27-32.

Ro, C. U., R. J. Vet, and D. Ord. 1998. Canadian Air and Precipitation Monitoring Network (CAPMoN) Annual Summary Reports (1983-1997). Environment Canada, Atmospheric Environment Service, Downsview, Ontario, Canada.

Schnoor, J. L. and W. Stumm. 1986. The role of chemical weathering in the neutralization of acidic deposition, Schweiz. Z. Hydrol., 48, 171-195.

Shaw, R. W. 1979. Acid precipitation in Atlantic Canada, Environ. Sci. Technol., 13, 406-411.

Sirois, A. 1993. Temporal variation of sulphate and nitrate concentration in precipitation in eastern North America: 1979-1990, Atmos. Environ., 27A, 945-963.

Stoddard, J. L., D. S. Jeffries, A. Lukewille, T. A. Clair, P. J. Dillon, C. T. Driscoll, M. Forsius, M. Johannessen, J. S. Kahl, J. H. Kellogg, A. Kemp, J. Mannio, D. Monteith, P. S. Murdoch, S. Patrick, A. Rebsdorf, B. L. SkjelkvŒle, M. P. Stainton, T. Traaen, H. van Dam, K. E. Webster, J. Wieting, and A. Wilander. 1999. Regional trends in aquatic recovery from acidification in North America and Europe 1980-95, Nature, In press.

Stumm, W. and J. J. Morgan. 1996. Aquatic Chemistry: chemical equilibria and rates in natural waters. Third Edition , John Wiley and Sons, NY.

Watt, W. D. 1987. A summary of the impact of acid rain on Atlantic salmon (*Salmo salar*) in Canada, Water, Air, Soil Pollut., 35, 27-35.

Watt, W. D., D. Scott, and S. Ray. 1979. Acidification and other chemical changes in Halifax County lakes after 21 years, Limnol. Oceanogr., 24, 1154-1161.

Table 1. Deposition trend analysis results at the regional CAPMoN sites. NC denotes no change.

	H+		SO_4^{-2}		NO_3^-		base cations	
	84-97	89- 97	84-97	89- 97	84-97	89- 97	84-97	89- 97
Kejimkujik	NC	⇓	⇓	NC	NC	NC	NC	NC

Table 2. Regional trends in lake chemistry acidification variables for Nova Scotia. The bold data are this study's results. The unbolded are results from previous analyses.

	up	Down	no		up	down	no
H^+				sulfate (SO_4^{-2})			
1983-1991	1	3	34	1983-1991	13	1	24
1983-1994	0	1	44	1983-1994	0	6	39
1983-1997	1	1	34	1983-1997	0	16	20
1989-1997	9	2	53	1989-1997	0	54	10
ANC				base cations (C_b)			
1983-1991	13	1	24	1983-1991	3	2	33
1983-1994	8	2	35	1983-1994	1	17	33
1983-1997	5	3	28	1983-1997	4	4	28
1989-1997	3	6	55	1989-1997	1	15	48

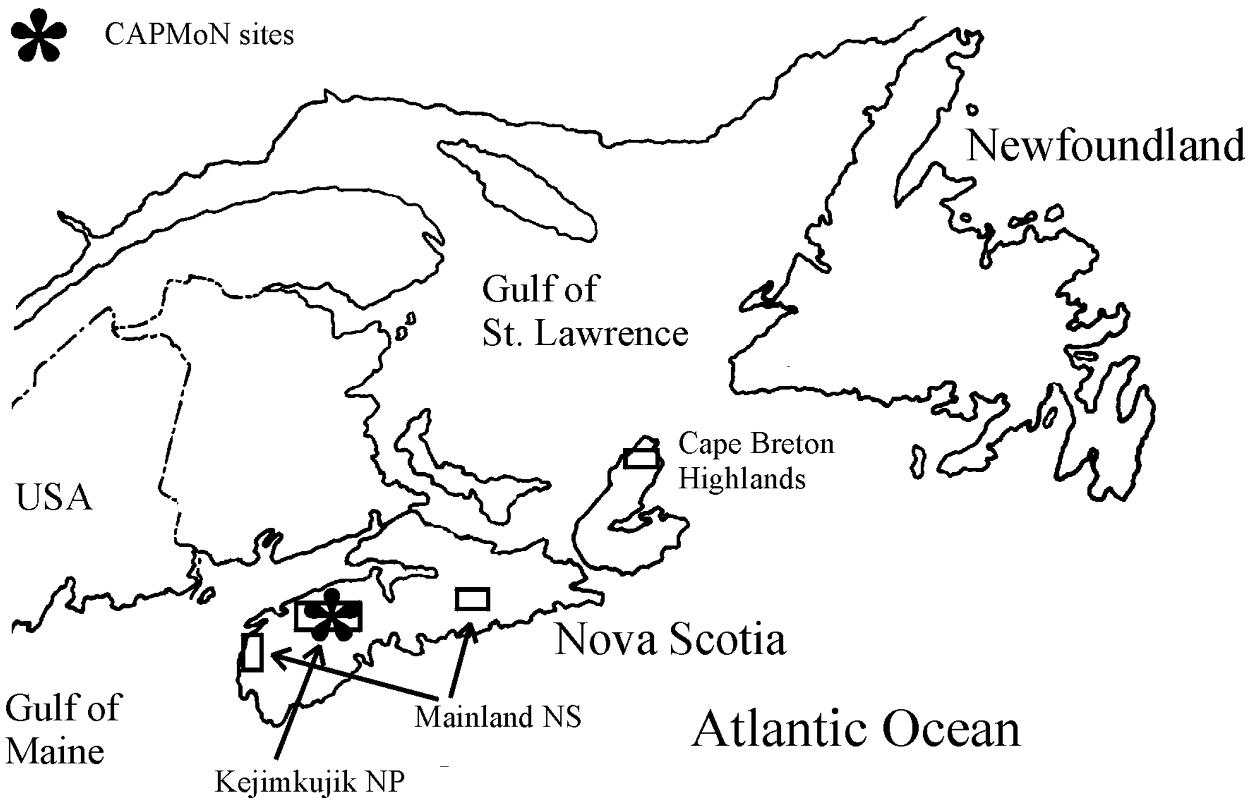

Fig. 1. Study lake locations.

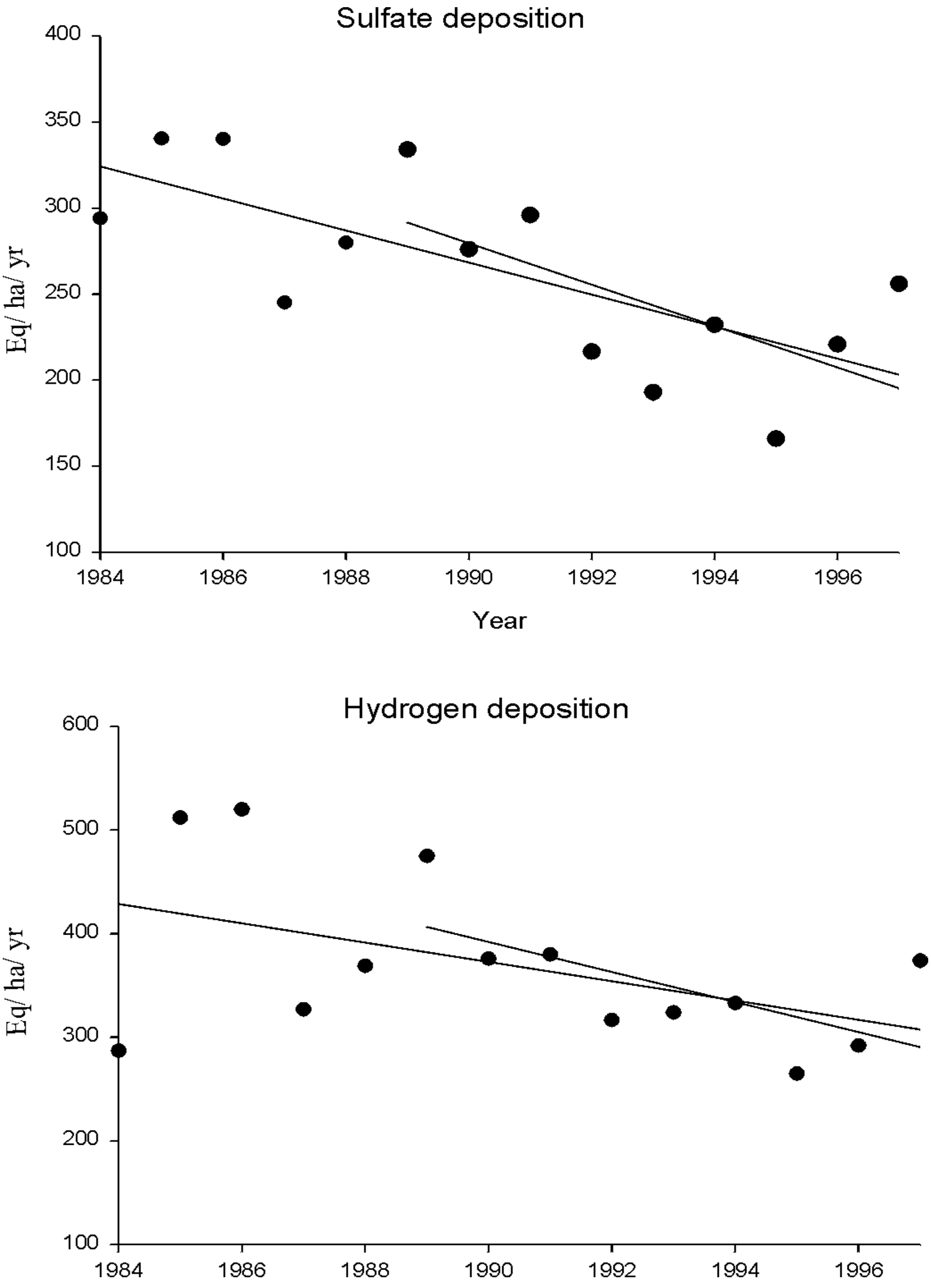

Fig. 2. Hydrogen and sulfate deposition at Kejimkujik NP. The regression lines are for the 1982-97 and 1989-97 periods.

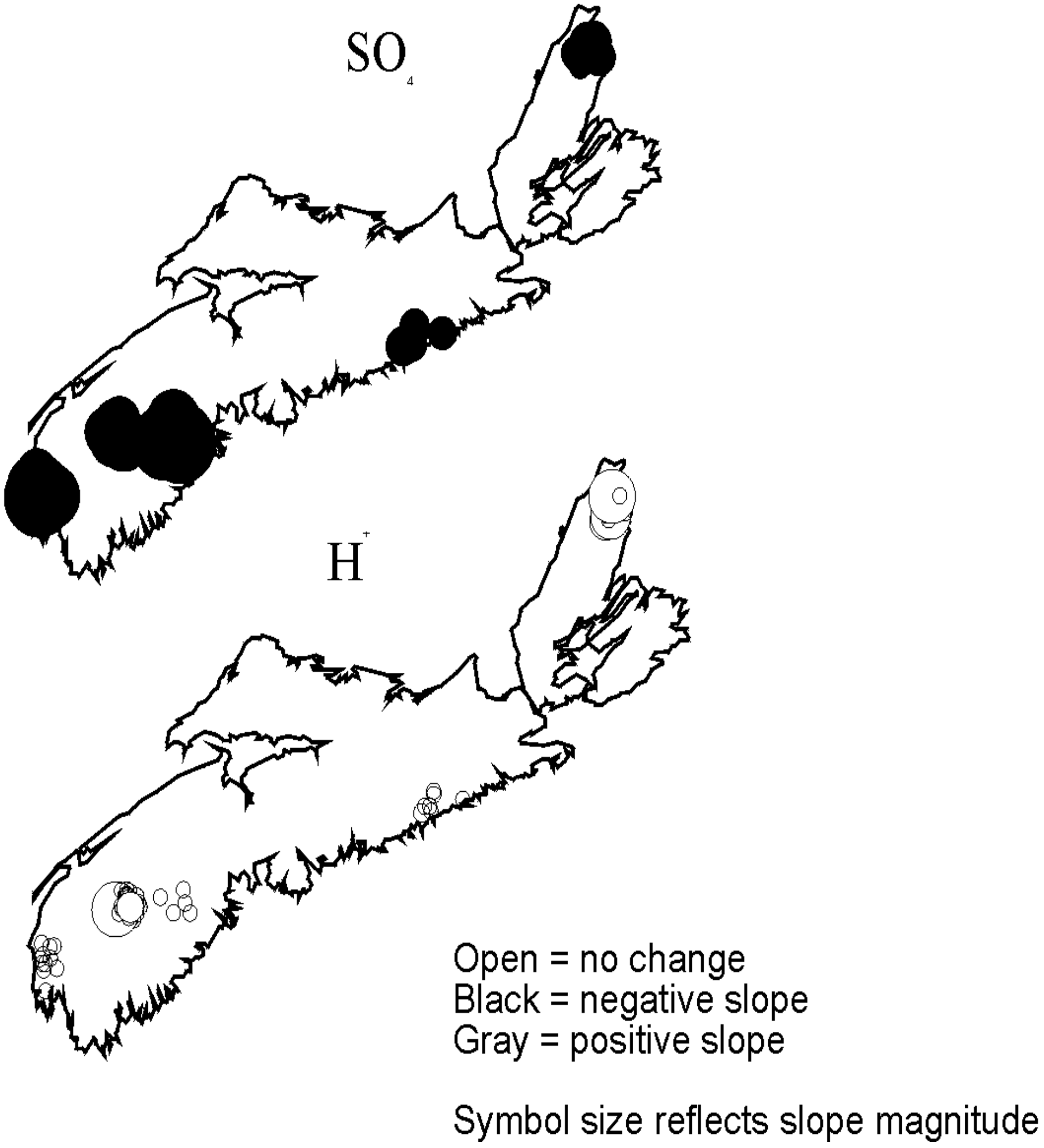

Fig. 3. Distribution of SO_4 and H^+ trends from 1989-97. The slopes are calculated using linear regression and are reflected in the relative size of the circles.

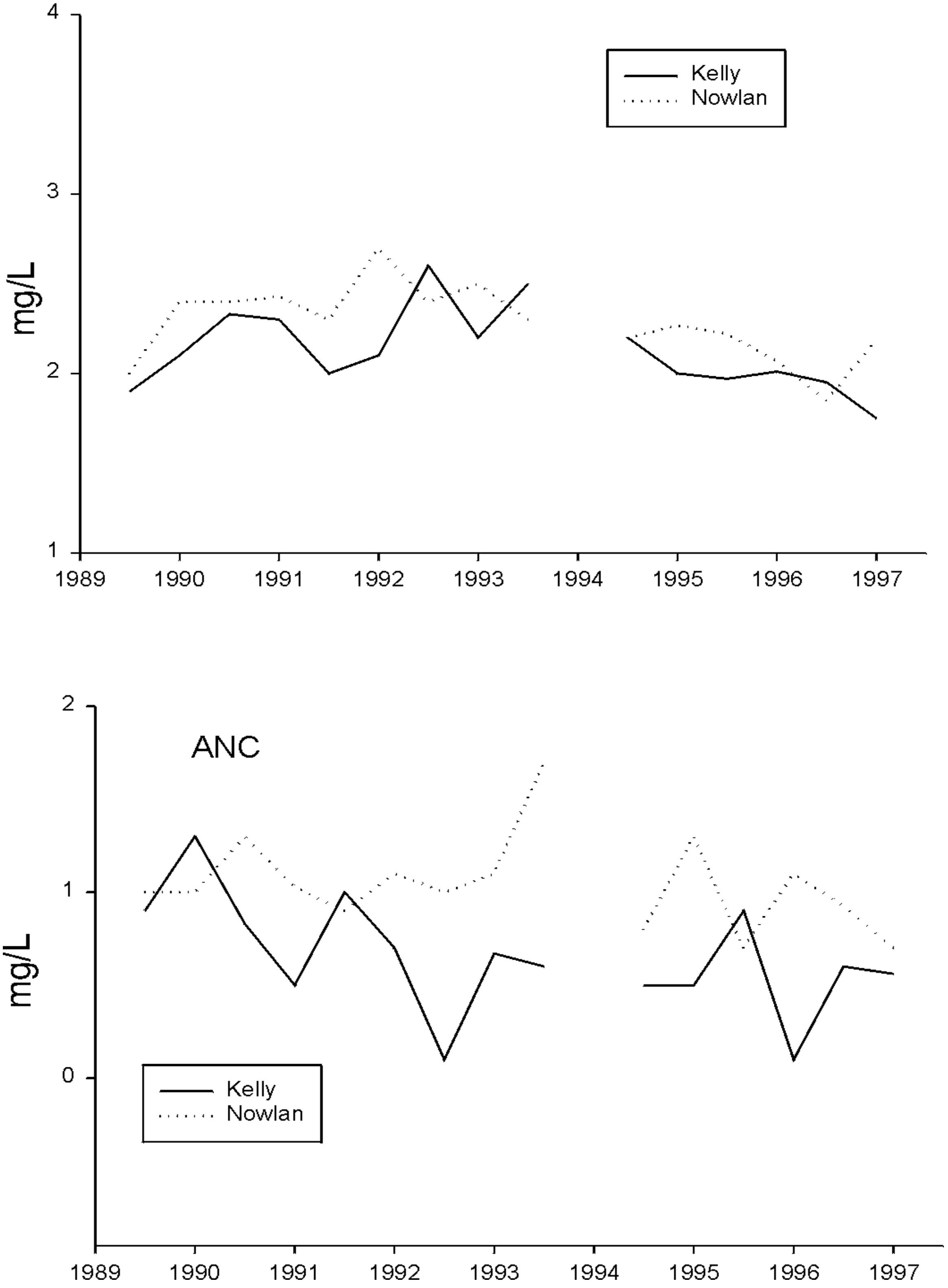

Fig. 4. Typical lake chemistry changes for SO_4 and ANC. (There were no samples collected in the fall of 1993).

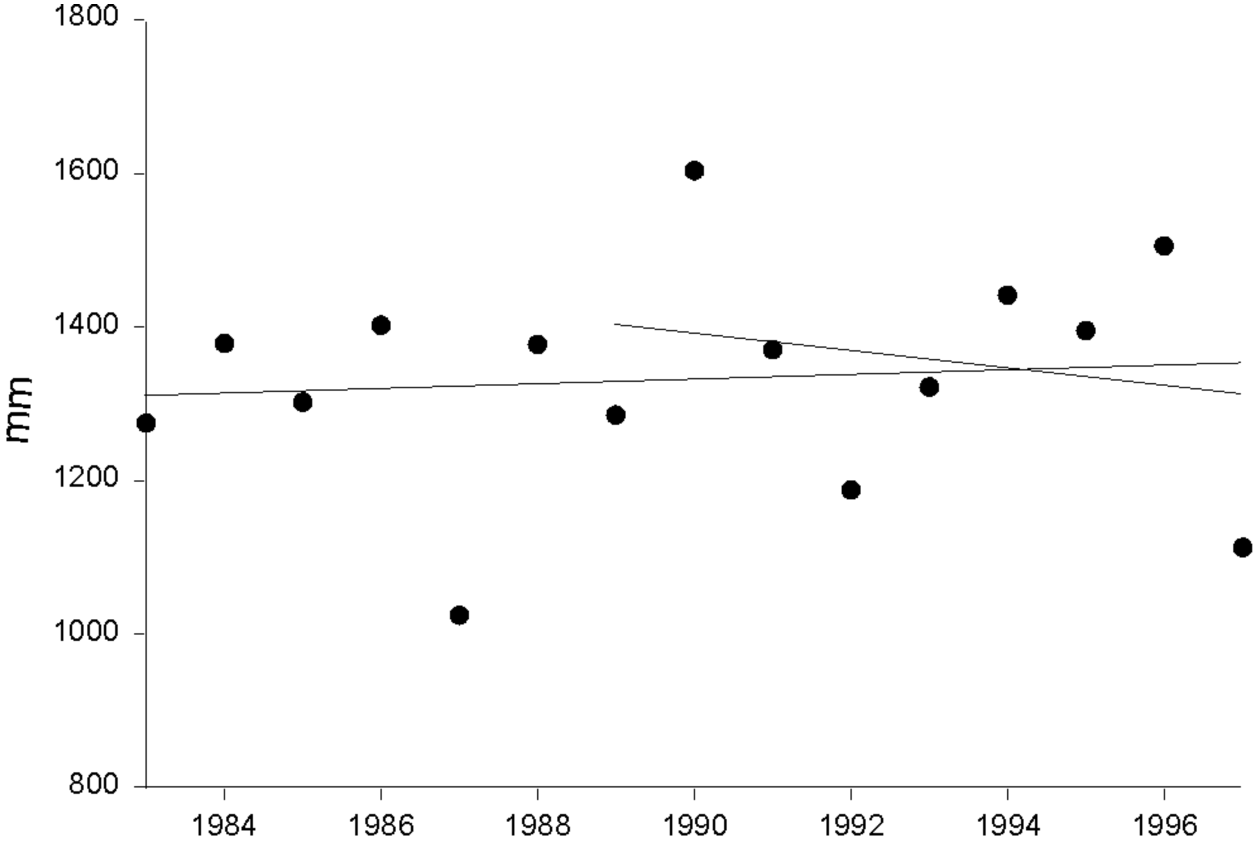

Fig. 5. Total annual precipitation at Kejimkujik. The regression lines are for the 1982-97 and 1989-97 periods.