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## Atlantic Region / Région de l'Atlantique





Environment Canada Atlantic Region

## Acid Precipitation during 1993 at Kejimkujik, N.S.

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## Abstract

The average precipitation-weighted pH for 1993 at Kejimkujik National Park in southwestern Nova Scotia was 4.66, which is 21% less acidic than the mean pH of 4.56 over the 14 years of record (1980-1993). The most acidic event of the year (pH of 3.13) was about 200 times more acidic than the least acidic event (pH of 5.44). Highly acidic precipitation events generally had air trajectories that crossed the more heavily industrialized sectors of the United States and Canada before arriving at Kejimkujik.

Wet deposition of excess sulphate at Kejimkujik during 1993 was 9.6 kg ha<sup>-1</sup>, the lowest amount measured at this site over its 14 years of record. The 1993 deposition was 34% below the 14-year mean of 14.6 kg ha<sup>-1</sup>. Wet deposition of nitrate during 1993 at Kejimkujik was 9.2 kg ha<sup>-1</sup>, which was 7% less than the 14-year mean of 9.9 kg ha<sup>-1</sup>. The largest daily deposition during 1993 occurred on June 18, when 0.4 kg ha<sup>-1</sup> of excess sulphate (over 4% of the annual total) and 0.06 kg ha<sup>-1</sup> of nitrate (3% of the annual total) were deposited.

A comparison of the average deposition for the first five years of the period of record to the average deposition for the most recent five years of the period showed that the average annual excess sulphate deposition decreased by almost 16%, whereas the average annual nitrate deposition increased by over 31%. An examination of the average yearly precipitation amount showed that there was little change (a 4% increase) from the first five years to the last five years.

Average precipitation-weighted concentrations for 1993 were 0.65 mg·l<sup>-1</sup> for excess sulphate and 0.14 mg·l<sup>-1</sup> for nitrate (expressed as nitrogen). The concentration of excess sulphate in precipitation has decreased since the early 1980's. The average precipitation-weighted concentration for the five years 1980 to 1984 was 1.16 mg·l<sup>-1</sup>, whereas the average concentration for the five year period 1989 to 1993 was 0.94 mg·l<sup>-1</sup>, representing a reduction of about 19%. The concentration of nitrate has generally been increasing in recent years. Average precipitation-weighted nitrate (as nitrogen) concentration for the five years 1980 to 1984 was 0.14 mg·l<sup>-1</sup>, whereas the average concentration for the five year period 1989 to 1993 was 0.18 mg·l<sup>-1</sup>, representing an increase of about 26%.

## 1. Introduction

Precipitation in Atlantic Canada is more acidic than in pristine areas of the world due mainly to the acidifying effect of anthropogenic sulphur dioxide and nitrogen oxide emissions. These gases are products of combustion in, for example, power plants, smelters, vehicles, residential furnaces and industrial processes in the areas upwind of the region. They are transformed into sulphuric  $(H_2SO_4)$  and nitric  $(HNO_3)$  acids in the atmosphere as they travel downwind from their sources, and they show up in precipitation as sulphates  $(SO_4^{-})$ , nitrates  $(NO_3^{-})$  and hydrogen ions  $(H^+)$ .

The pH value of precipitation is a convenient measure of the hydrogen ion concentration (acidity) of the precipitation. A pH value of 5.6 is the characteristic acidity of precipitation which has no pollutants. Precipitation which has a pH lower than 5.6 (more acidic) is called "acid precipitation". Most of eastern Canada receives precipitation with average annual pH values ranging from 4.2 to 4.5.

Different landscapes have different acid buffering capacities and, therefore, differing sensitivities to the effects of acid precipitation. Those with low alkalinity and, therefore, low buffering capacity are especially sensitive to acid precipitation.

Damage to lakes and streams is usually observed in acid sensitive areas that regularly receive precipitation with pH less than 4.7. Readings of 4.2 and below are considered strongly acidic and readings of less than 4.0 are considered to be serious events, although they do not constitute an immediate danger to human health or property. The effects of acid rain and snow are generally cumulative over time, although fish kills have been observed after low pH events and after the melting of acidic snow (RMCC, 1990).

Another measure of the potential for environmental damage is the amount of acidic loading that occurs. This is measured in terms of the amount of acid that falls from the atmosphere per unit area (the acidic deposition) over a given time period. Deposition of acid from the atmosphere occurs continuously, both during and between precipitation events. The former deposition is called "wet" deposition and the latter is referred to as "dry" deposition.

Wet excess (non-sea salt) sulphate deposition ("excess" because the contribution from sea salt has been deducted) is used as a surrogate for the total anthropogenic acid deposition because anthropogenic sulphuric acid usually dominates the acidification process (MOI, 1983). However, another source of non-anthropogenic sulphate, that of biogenic emissions from the ocean, is not considered when calculating excess sulphate. Although data are too limited to accurately determine the magnitude of the marine biogenic contribution to sulphate loadings, it is estimated that anywhere from a negligible component (Wadleigh, 1994) to approximately 20% of the excess sulphate at Kejimkujik (Beattie et 1990) is of marine biogenic origin, al., rather than of anthropogenic origin. Further study is continuing to assess the biogenic component.

An analysis of over a decade of Canadian acid rain research has led to the conclusion that the critical load for most of Atlantic Canada (with the exception of PEI, which is well buffered) is less than 8 kg ha<sup>-1</sup> yr<sup>-1</sup> of excess sulphate from precipitation (RMCC, 1990). This critical load is exceeded in the very sensitive areas of southern Nova Scotia and New Brunswick, even at locations remote from local pollution sources, due to acidifying pollutants from upwind regions.

In response to the initial concern about the potentially adverse effects of acidic deposition in Atlantic Canada, the Atmospheric Environment Service (AES) of Environment Canada began operating a daily precipitation sampling station in Kejimkujik National Park in southwestern Nova Scotia in May 1979. This site is located in an acid sensitive area downwind from the major pollution sources in North America (Figure 1) and regularly receives precipitation with pH less than 4.7. The precipitation samples collected at the site are analyzed for pH (acidity) and for the concentration of various chemical constituents, including sulphates and nitrates, the major acidifying components of acid The results are available in AES data reports; precipitation. prior to June 1983, the APN (Air and Precipitation Monitoring Network) reports and, presently, the CAPMON (Canadian Air and Precipitation Monitoring Network) reports.

In order to provide information on precipitation acidity more quickly, AES issued a weekly bulletin, called the Acid Rain Report, for a period of over 11 years (December 1983 to March 1995). This bulletin listed the preliminary pH ("field pH") measurements, and the precipitation amounts and types, for the precipitation events that occurred during the previous week at sites in eastern Canada, including Kejimkujik. The bulletin also included a list of areas over which the air had passed before arriving at the collection site (the "air path to site"), thus giving information on the source of the pollutants associated with these precipitation

Analyses of the CAPMON data collected at Kejimkujik have been prepared on a frequent basis. Recent analyses of trends for the period of record are contained in Sirois (1993) and Beattie and Keddy (1993). More detailed analyses for individual years for 1986 to 1992, respectively, are contained in: Pettipas and Beattie (1987), Allen and Beattie (1988), Hanley and Beattie (1989), Beattie *et al.* (1990), Beattie and O'Brien (1992a and 1992b), and Beattie *et al.* (1994).

The objectives of this report are: (1) to characterize the nature and variations of the concentration of sulphate, nitrate and hydrogen ions in precipitation and their deposition amounts during 1993 at Kejimkujik National Park; (2) to investigate the relationship between pollutant sources and resulting precipitation acidity at Kejimkujik; and (3) to compare the 1993 precipitation chemistry data with data from previous years.

## Figure 1

Location of Monitoring Site at Kejimkujik National Park, Nova Scotia, in Relation to Pollution Sources and Storm Tracks



Mean monthly tracks of frontal storms (1885-1980) in eastern North America. Sixty-five percent of all sulphur dioxide and 49 percent of all nitrogen oxides produced in North America are emitted in the dark area to the west of New England. (Bormann, 1982).

### 2. Methods

### 2.1 Precipitation Chemistry Measurement

The location of the Kejimkujik station was chosen to avoid any local pollution sources and to minimize local site influences on precipitation quality and quantity, thus providing regionally representative results (Vet *et al.*, 1986). Precipitation is sampled using an automated collector that uncovers the collection bucket during periods of precipitation. The sample bags are removed on a daily basis according to a strict protocol that was designed to avoid contamination of the samples, as outlined in Vet *et al.* (1989). The samples are analyzed for ionic constituents at a dedicated precipitation chemistry laboratory, usually within 10 days of receipt. The analytical methods, techniques, and quality control and assurance procedures are outlined in Vet *et al.* (1989).

The precipitation amount is measured as a depth using an AES standard rain gauge and standard Nipher-shielded snow gauge, which are collocated with the precipitation chemistry collector. The snow is melted and measured as a water equivalent. The precipitation type is determined by the site operator based on direct evidence or weather reports.

## 2.2 Precipitation Chemistry Data Preparation

The daily pH, hydrogen ion concentration ([H<sup>+</sup>]), excess (nonsea salt) sulphate concentration ([XSO4<sup>\*</sup>]), nitrate (expressed as nitrogen) concentration  $([NO_3^--N])$ , and precipitation type and amount were extracted from the Kejimkujik CAPMON quality-controlled precipitation chemistry data set for 1993 (Appendix B). The H<sup>+</sup> and XSO4 concentrations were derived from measured parameters, as outlined in Vet et al. (1989): i.e., the concentration of H<sup>+</sup> was calculated from the measured pH; the concentration of excess sulphate was determined by subtracting the sulphate contributed by sea salt from the measured sulphate in the sample. The concentration of sea salt sulphate in each sample was estimated from the concentration of either sodium, magnesium or chloride, depending on which one appeared to be the best indicator of sea salt in the sample (see Vet et al. (1989) for details of the This correction is recommended for sites located procedure). within 100 km of the coast (UDDBC, 1985).

In preparing the data set, valid concentration and precipitation data designated as non-detectable were set equal to one-half the non-detectable value for calculation purposes (Vet *et al.*, 1989), but are retained in their original form (preceded by a "minus" designator) in the listing in Appendix B.

It was found that 10 out of the 218 precipitation days had insufficient precipitation amounts for any chemical analysis. These entries were eliminated from the data set. Of the remaining 208 precipitation days, 3 were rejected as invalid due to possible contamination (the collector was open when precipitation was not occurring), representing a precipitation amount of 37.2 mm. Twenty-seven additional samples in the data set, although not rejected, had missing pH measurements (precipitation total 10.6 mm). These missing pH values are designated by -9's in Appendix B.

The final precipitation chemistry data set for 1993 (Appendix B) thus contained 205 precipitation days (for a total of 1445.5 mm or 97% of the annual total of 1484.2 mm), of which 178 had pH and  $[H^+]$  values, and the full 205 had XSO<sub>4</sub><sup>=</sup> and NO<sub>3</sub><sup>-</sup>-N concentrations.

Due to the importance of wet deposition to the acidification process, the concentrations of  $H^*$ ,  $XSO_4^{=}$  and  $NO_3^{-}-N$  in each precipitation sample were converted into deposition amounts (in mg·m<sup>-2</sup>) by multiplying the chemical concentration (in mg·l<sup>-1</sup>) by the amount of precipitation (in mm) for each precipitation event. (Units of kg·ha<sup>-1</sup> for wet deposition can be obtained from mg·m<sup>-2</sup> by dividing by 100.)

To identify the pH class that made the dominant contribution to acidic deposition, the daily H<sup>\*</sup> depositions for the 178 days with valid pH measurements were summed for four pH classes (pH > 5.6;  $4.7 < pH \le 5.6$ ;  $4.0 < pH \le 4.7$ ; pH  $\le 4.0$ ).

For each chemical, the daily deposition amounts were summed for the year, by season, and for different types of precipitation, due to missing concentration data, this produced but an underestimate of the actual deposition amounts in each case. Therefore, the sums were converted into average precipitationweighted concentrations, and then multiplied by the total precipitation for the appropriate summary period or type of precipitation to give a better estimate of the actual deposition amounts, based on the procedures outlined in UDDBC (1985) and Vet Following conventional practice, the average *et al.* (1989). nitrate concentrations were expressed as nitrogen concentrations (i.e.  $[NO_3^- - N]$ ), whereas wet deposition was expressed in terms of the complete ion (i.e.  $NO_3^{-}$ ). The conversion factor from  $NO_3^{-}-N$  to  $NO_3$  is 4.43 (the ratio of the molecular weight of  $NO_3$  to the atomic weight of N).

Average pH values were calculated from the average precipitation-weighted hydrogen ion concentrations, according to the procedure used by Vet *et al.* (1989).

To compare the relative contribution of excess sulphate and nitrate to precipitation acidity, annual and seasonal depositions were converted into ion (chemical) equivalents (in meq<sup>-2</sup>) by multiplying the deposition (in mg<sup>-2</sup>) by the ion valence and then dividing by the molecular weight.<sup>1</sup> Thus, the ratio of excess sulphate deposition to nitrate deposition was converted into its ion equivalent ratio by multiplying by 1.29.

<sup>&</sup>lt;sup>1</sup> The valence for the sulphate ion is 2 and for the nitrate ion, one; the molecular weight for sulphate is 96.1 g mole<sup>-1</sup> and for nitrate, 62.0 g mole<sup>-1</sup>.

## 2.3 Determination of Pollutant Source Areas

The "air path to site" information in the Acid Rain Report was determined from an analysis of weather patterns and by using a three-dimensional back-trajectory model (Olson *et al.*, 1978). This model calculates the air parcel's previous positions during the past 120 hours using the wind and other meteorological data at various heights above ground, usually at the 1000, 925 and 850 mb levels in the lower atmosphere (Desautels, 1985), which correspond to heights just above the ground, about 700 m above the ground, and about 1500 m above the ground, respectively.

The trajectory information for Kejimkujik was available for 134 precipitation days in 1993 (representing 1401.1 mm of precipitation or 94% of the annual total). All but two of the days without this information had small precipitation amounts (less than 1.0 mm). However, no trajectory information was available for Apr. 30 and May 1, when 13.0 mm and 4.0 mm of precipitation fell, respectively.

The trajectory information was incorporated into the precipitation chemistry data set by first dividing eastern North America into four sectors, centred on Kejimkujik (Figure 2), and then, for each precipitation event, identifying which sector(s) the trajectory had crossed before arriving at Kejimkujik. The sectors were numbered in ascending order based on the amount of anthropogenic emissions of sulphur oxides  $(SO_x)$  and nitrogen oxides  $(NO_x)$  in 1985 (most recent data available), as shown in Table 1.

To investigate the relationship between upwind emission levels and precipitation acidity at Kejimkujik, the assumption was made that there was one major source sector of pollutants for each precipitation event (Desautels, 1986; Pettipas and Beattie, 1987; Beattie and O'Brien, 1992a). Of the 178 days during 1993 with precipitation pH values, 45 did not have corresponding trajectory information and therefore, the upwind sectors were unknown. Of the remaining 133 days, the upwind source was unambiguous for 89 days (the trajectory to Kejimkujik crossed only one sector), but was ambiquous for the remaining 44 days (the trajectory crossed two sectors). In these 44 cases, the upwind sector with the highest sulphur oxide emissions (see Table 1) was assumed to be the major source sector, unless the trajectory only touched the sector. Of the 205 days in 1993 with  $XSO_4^{-}$  and  $NO_3^{-}-N$  concentrations, 71 did not have corresponding trajectory information. Of the remaining 134 days, the upwind source was unambiguous for 89 days, and ambiguous for 45.



Four Sectors Surrounding Kejimkujik, N.S.

Figure 2

		• ·	
Sector	Description	Em: SO <sub>x</sub> (ki:	issions NO <sub>x</sub> lotonnes)
I	Atlantic Ocean	-	~
II	N.S., P.E.I., N.B., Nfld, Labrador, Nrn Quebec	414	183
III	Srn and Wrn Quebec, Nrn Ontario, Nrn and Central Maine, Minnesota, Nrn Michigan, Wisconsin	2536	1226
IV	Srn Ontario, all of USA East of Dakota-Texas line (except that in Sector III)	16090	11888

Table 1. 1985 Anthropogenic Emissions of  $SO_x$  and  $NO_x$  from Sectors Surrounding Kejimkujik, N.S.

Source of data: RMCC, 1990

### 3. Results and Discussion

### 3.1 Precipitation Event Characteristics

The considerable variability in daily  $H^*$ ,  $XSO_4^*$  and  $NO_3^-N$  concentration and deposition amounts during the year can be seen from the CAPMON Summary Statistics (Appendix A) and the data listings (Appendix B).

An analysis of the distribution of pH in precipitation events (Figure 3) shows that all of the precipitation events with valid pH data at Kejimkujik in 1993 were acidic (pH  $\leq$  5.6). Almost half were within the moderately acidic range (4.0 < pH  $\leq$  4.7) and a significant number were in the highly acidic pH range (pH  $\leq$  4.0). Table 2 summarizes these events. The most acidic event of the year had a pH of 3.13 (October 7), which was 204 times more acidic than the least acidic event (pH 5.44 on January 27).



•

· · · · · · · · · · · · · · · · · · ·		Number of	
Level of pH	pH Range	Occurrences	<u>Percentage</u>
Normal (clean) Slightly Acidic Moderately Acidic Highly Acidic	$\begin{array}{rrrr} pH > 5.6 \\ 4.7 < pH \leq 5.6 \\ 4.0 < pH \leq 4.7 \\ pH \leq 4.0 \end{array}$	0 62 86 30	0% 35% 48% 17%
	Total:	178	100%

Table 2:	Summary of Pr	ecipitation	pH Di	istribution
	at Kejimkujik	, N.S. Durin	ig 199	93

Very low pH values (high acidity) and high concentrations of acidifying chemicals are generally associated with events that produce only a small amount of precipitation (Anlauf *et al.*, 1975). Therefore, the total acid deposition from these events may not be as large as for other, higher pH events with more precipitation. Plots of the daily concentration of hydrogen ion, excess sulphate and nitrate versus daily precipitation amount (Figures 4 to 6, respectively) graphically illustrate the usual relation between concentration and precipitation amount, and the effect on daily deposition amounts; i.e. the largest depositions usually occur for events with moderate concentrations and moderate precipitation amounts.

## 3.2 Concentration and Deposition by Precipitation Type

The three different types of precipitation that were recorded at Kejimkujik during 1993 (rain, snow, and mixed rain and snow) accounted for significantly different amounts of the total deposition of pollutants during the year. This was largely a consequence of the predominance of rain at Kejimkujik. Of the total of 218 days with precipitation at Kejimkujik during 1993 (total amount 1484.2 mm), rain occurred on 146 days (total amount 1003.2 mm), snow on 61 days (total amount 287.0 mm water equivalent), and mixed rain and snow occurred on 11 days (total amount 194.0 mm water equivalent).

The analysis for pH and  $H^{+}$  deposition by precipitation type is shown in Table 3. Rain events had the highest deposition (78% of the total). On average, during 1993, as in 1992 and in three of the previous six years, rain was somewhat more acidic than snow (lower average pH).

Relation between H+ Concentration and Daily Precipitation Amount Kejimkujik, N.S. 1993 Figure 4





Relation between Nitrate (NO3-N) Concentration and Daily Precipitation Amount Kejimkujik, N.S. 1993 Figure 6



Table 3:	Average pH	and H	I⁺ Dep	osition	by	Precipitation	Type
	for Kejimk	ujik,	N.S.	During	1993		

Precipitation Type	Average pH	Deposition of H <sup>+</sup> (kg <sup>·</sup> ha <sup>-1</sup> )	Percentage Deposition
Rain	4.60	0.251	78%
Snow	4.76	0.050	15%
Mixed	4.93	0.023	7%

The analysis for  $XSO_4^{\dagger}$  and  $NO_3^{-}$  deposition is shown in Table 4. Rain events, not surprisingly, had the highest deposition (82% of annual deposition for excess sulphate and 73% of annual deposition for nitrate). Snow events had, on average, considerably lower excess sulphate concentrations than rain events, but higher nitrate concentrations.

Table 4: XSO4<sup>\*</sup> and NO3<sup>-</sup> Precipitation-weighted Average Concentrations (mg·l<sup>-1</sup>) and Deposition Amounts (kg·ha<sup>-1</sup>) by Precipitation Type for Kejimkujik, N.S. During 1993 (% of Annual Deposition in Brackets)

Precipitation Type	Conc.	XSO4 Depo	sition		NO3 <sup>-</sup> -N Conc.	N Depo	N <sub>3</sub> sition
Rain	0.79	7.9	(82%)	(1)	0.15	6.7	(73%)
Snow	0.36	1.0	(11%)		0.16	2.0	(22%)
Mixed	0.38	0.7	(8%)		0.05	0.4	(5%)

NOTE:

(1) Percentages do not total 100% due to rounding.

#### 3.3 Seasonal and Annual Summary

The seasonal characteristics of precipitation acidity were investigated by grouping the data into three-month periods: January to March (Winter), April to June (Spring), July to September (Summer) and October to December (Fall). Table 5 shows the number of precipitation days, the total precipitation, the average precipitation-weighted pH and the concentration and deposition of H<sup>+</sup>, excess sulphate and nitrate for each season and for the year. Due to the procedure used to compensate for missing data, the seasonal depositions (Table 5) and the precipitation type depositions (Tables 3 and 4) may not sum to the annual depositions (Table 5).

	Pre	and fo	the Year at Average	Kejimku Concen	jik, N.S. Du tration	cing 1993 Wet D	enositior	(1)	
Period	Days	Amount (mm)	Hđ	XSO4	NO <sub>3</sub> <sup>-</sup> - N	H <sup>+</sup>	XSO4	NO3-	
Winter	67	423.8	4.86	0.35	0.10	0.06	1.5	1.9	
Spring	54	313.4	4.51	1.07	0.18	0.10	3.4	2.5	
Summer	40	188.3	4.41	1.14	0.21	0.07	2.1	1.8	
Fall	57	558.7	4.78	0.45	0.12	0.09	2.5	3.0	
Annual	218	1484.2	4.66	0.65	0.14	0.32	9.6	9.2	
NOTE :									

Average Precipitation-weighted pH, Concentration (mgl<sup>-1</sup>)

Table 5:

(1) Annual totals may not equal the sum of seasonal values due to rounding and/or procedure used to compensate for missing data.

15

According to Table 5, summer had the lowest average pH, but since it had by far the least precipitation of any season, the largest deposition of H<sup>+</sup> actually occurred in spring. Winter had the highest average pH. It is unusual for the lowest average pH to occur in summer at Kejimkujik; spring usually has the lowest pH, as in 1991 and 1992 (Beattie *et al.*, 1994; Beattie and O'Brien, 1992b), and 1986 through 1989 (Beattie *et al.*, 1990; Hanley and Beattie, 1989; Allen and Beattie, 1988; and Pettipas and Beattie, 1987). However, in 1990, the average summer pH was considerably lower than spring, although the fall pH was the lowest (Beattie and O'Brien, 1992a).

Spring, 1993 had the highest deposition of excess sulphate, while for nitrate, the largest deposition occurred during the fall. The highest average precipitation-weighted concentrations (wet deposition divided by precipitation amount) for all three ions occurred during the summer.

Although the average annual pH at Kejimkujik in 1993 (4.66) was considerably higher than that for 1992 (4.57), deposition totals of  $H^+$  (0.32 kg ha<sup>-1</sup>) were the same both years. Deposition of  $XSO_4$  (9.6 kg ha<sup>-1</sup> in 1993 compared to 10.8 in 1992) and  $NO_3$  (9.2 kg  $ha^{-1}$  in 1993 compared to 9.4 in 1992) were lower in 1993. Lower concentrations of all three chemicals in 1993 were at least partially offset by a greater precipitation amount (1484.2 mm compared to 1176.7 in 1992, or an increase of 26%). To illustrate this point in more detail, the annual H<sup>+</sup> deposition at Kejimkujik in 1993 (32.4 mg m<sup>-2</sup> or 0.32 kg ha<sup>-1</sup>) represented a slight increase of 2% from the 1992 deposition of 31.7 mg m<sup>-2</sup>. However, the precipitation-weighted concentration of H<sup>+</sup> was only 0.0218 mg l<sup>-1</sup> in 1993, compared to 0.0269 mg  $l^{-1}$  in 1992, representing a 19% decrease (A.E.S., 1993a). (Since the normal amount of precipitation at Kejimkujik National Park (based on 1966-1990) is 1396.9 mm (A.E.S., 1993b), the precipitation in 1993 was just 6% above normal.)

Seasonal ratios of excess sulphate to nitrate deposition based on ion equivalents (Figure 7) were found to be higher in the spring and summer seasons (April to September) and lower in the fall and winter seasons (October to March). These ratios indicate that, on an ion equivalent basis, excess sulphate contributed almost twice as much to the acidity of the spring and summer deposition as nitrate; whereas, in the fall and winter, nitrate contributed about the same amount as excess sulphate. Similar results were found in earlier studies: for the period 1979-1987 (Sirois and Summers, 1989), for 1990-1992 (Beattie et al., 1994; Beattie and O'Brien, 1992a; 1992b) and for Harcourt, N.B., in 1991 (Beattie et al., 1993). Harcourt is also a remote site at about the same longitude as Kejimkujik, but two degrees farther north.



## 3.4 Source Contributions to Precipitation Acidity

The results of the analysis of source contributions to precipitation acidity are plotted in a multiple pie chart in Figure These pies illustrate the air mass back-trajectory information 8. for 1993 (as described in section 2.3), and show the controlling influence of the major storm tracks which bring precipitation into the Maritimes. As shown in Figure 1 (from Bormann, 1982), one major storm track lies along the U.S. East Coast, while another track, really an amalgamation of the Colorado and Alberta storm tracks, crosses the Great Lakes, southern Ontario, southern Quebec and Maine before it reaches the Maritimes. Each pie in Figure 8 represents a pH range and each slice represents a different source sector (as defined in Table 1 and Figure 2). The data printed immediately after each sector label represent the percentage of precipitation events (with the concentration in the range indicated) for which that sector was the "major" source sector (as described in Section 2.3). The total number of precipitation events with valid measurements is printed below each pie.

Figure 8 illustrates, for example, that precipitation events Kejimkujik with pH in the highly acidic range (pH  $\leq$  4.0) at generally had air mass trajectories which crossed the more heavily industrialized regions in the United States and Canada (Sector IV). This was the major source sector for 92% (12 out of 13) of the events with known source regions. Precipitation events with less acidic levels show a reduced influence from Sector IV (southern Ontario and the U.S.). For instance, the trajectories to Kejimkujik had Sector IV as the major source sector in 48% (31 out of 64) of the events with known source regions which had pH in the moderately acidic range (4.0 < pH ≤ 4.7). Precipitation events with only slightly acidic pH values (pH > 4.7) predominantly had Atlantic Ocean (Sector I) as the major source sector. the Moreover, only 32% (18 out of 56) of the events with known trajectories which had precipitation pH in the slightly acidic range had Sector IV as the major source sector.

Further confirmation of the influence of upwind emission levels on the precipitation chemistry at Kejimkujik can be seen by examining the average precipitation-weighted pH,  $XSO_4$  and  $NO_3$  -N concentrations measured in precipitation with air trajectories from each major source sector (Table 6). For the most part, the average decreased and the average excess sulphate and nitrate Hq concentrations increased as the sector emission levels increased (see Table 1). However, rather surprisingly, Sector III had higher average concentrations of acidifying chemicals than Sector IV. An examination of precipitation events showed a much smaller number of events arriving from Sector III, with generally low precipitation tendency for low-amount events to have higher amounts. The concentrations would likely account for part of the reason why Sector III events were more polluted; moreover, the sample size was very small (20 events). (Depositions, on the other hand were four to seven times greater with trajectories from Sector IV than from Sector III, depending on the chemical.)



		рН	[XSO <sub>4</sub> =] mg·l <sup>-1</sup>	[NO <sub>3</sub> -N] mg·l <sup>-1</sup>
Sector	I	5.00	0.30	0.044
Sector	II	4.86	0.36	0.066
Sector	III	4.40	0.99	0.368
Sector	IV	4.50	0.92	0.195

Table 6: Precipitation-weighted Average pH, and Average XSO<sub>4</sub><sup>=</sup> and NO<sub>3</sub><sup>-</sup>-N Concentrations in Precipitation by Major Source Sector at Kejimkujik, N.S., 1993

### 3.5 Episodic Contributions to Acidic Deposition

Of particular interest in the investigation of acidic deposition is the contribution of individual large deposition events, referred to as "episodes" (Pasquill and Smith, 1983). А study of large deposition episodes occurring at Kejimkujik during the early part of the data record (Beattie, 1987; Beattie and Whelpdale, 1989) showed that a large percentage of the annual wet deposition was deposited by only a small percentage of the precipitation events during the year. It is likely that this characteristic occurs every because the episodic year meteorological situations that produce these episodes almost always have the same features (a southwesterly or westerly wind flow transporting pollutants from the major source regions in the U.S., followed by the passage of a precipitation-bearing weather system generally arriving from the west).

The investigation for 1993 again showed this episodic characteristic. After ranking the 205 precipitation days in descending order based on the daily wet deposition amount for each chemical separately, it was found that 30% of the total measured wet deposition was contributed by 6 to 8% of the precipitation days.

For H<sup>+</sup> deposition, about 8% (14 out of 178) of the days with pH measurements were responsible for over 30% of the measured H<sup>+</sup> deposition for the year. For  $XSO_4^{\pm}$  and  $NO_3^{-}$ , about 6% (13 out of 205 days for each chemical) were responsible for over 30% of the measured wet deposition for each chemical. Therefore, the deposition at Kejimkujik in 1993 qualified as episodic for H<sup>+</sup>,  $XSO_4^{\pm}$  and  $NO_3^{-}$ , but not highly episodic (when less than 5% of the annual precipitation days are episode days). High episodicity usually occurs in areas remote from the largest source regions (Pasquill and Smith, 1983).

A seasonal analysis of the large deposition episodes showed a tendency to occur during the warmer seasons in 1993, especially for the hydrogen and sulphate ions. Less than 15% of the large deposition events occurred in the winter months (January to March), depending on the ion (0% for sulphate, 7% for hydrogen ion and 15% for nitrate). About an equal number of events occurred during the other three seasons. A spring - summer bias was found in the four years 1980-1983 (Beattie, 1987; Beattie and Whelpdale, 1989), but not in 1990 (Beattie and O'Brien, 1992a). This bias was again noted in 1991 and 1992 at Kejimkujik (Beattie and O'Brien, 1992b; Beattie *et al.*, 1994), as well as in 1991 at Harcourt, N.B. (Beattie *et al.*, 1993).

There were also biases in precipitation type, with 13 out of 14 (93%) of the large deposition episodes for hydrogen ion being rain events, while the remaining event consisted of snow. Twelve out of 13 (92%) of the large deposition events for excess sulphate were rain events, with one mixed event (consisting of both rain and snow). For nitrate, 11 of the 13 episodes (85%) were rain events, and the remaining two were snow events. Similar biases were found for the early 1980's (Beattie, 1987; Beattie and Whelpdale, 1989) and for 1990-1992 (Beattie and O'Brien, 1992a and 1992b; Beattie et al., 1993; Beattie et al., 1994).

Event #	Date M/D	Precip Type (1)	<u>pitation</u> Amount (mm)	Upwind Sectors (2)	рH	Wet H+ I Amount mg·m <sup>-2</sup>	Deposition % of Total (3)
1 2 3 4 5	06/28 06/18 08/20 06/21 12/21	R R R R R	12.4 2.5 11.6 4.8 37.0	IV IV IV IV,I IV,I	4.03 3.34 4.11 3.78 4.72	1.16 1.14 0.90 0.80 0.71	3.7% 3.7% 2.9% 2.6% 2.3%
				Total:		4.70	15.1% (4)

Table 7:	The Five Large	st Wet H <sup>+</sup>	Deposition	Events
	at Kejimkujik,	N.S. Dur	ing 1993	

#### NOTES:

(1) Precipitation Type: R = Rain

(2) Upwind sectors are defined in Table 1.

(3) Total measured deposition was  $31.2 \text{ mg m}^{-2}$ .

(4) Any difference between sum and Total is due to rounding.

Table	8:	The	Five	Larges	st Wet	XSO₄ <sup>≠</sup>	Deposition	Events
		at :	Kejimł	cujik,	N.S.	During	1993	

		Preci	pitation		<u> </u>	Wet XSO	4 Deposition
Event #	Date M/D	Type (1)	Amount (mm)	Upwind Sectors (2)	[XSO4=] mg·l-1	Amount mg·m <sup>-2</sup>	% of Total (3)
1	06/18	R	2.5	IV	16.35	40.9	4.4%
2	06/28	R	12.4	IV	2.54	31.5	3.4%
3	08/20	R	11.6	IV	2.37	27.5	2.9%
4	06/21	R	4.8	IV,I	5.67	27.2	2.9%
5	05/16	R	3.8	IV	6.28	23.9	2.5%
				Total:		150.9	16.1% (4)

NOTES:

Precipitation Type: R = Rain
 Upwind sectors are defined in Table 1.

(3) Total measured deposition was 937.9 mg·m<sup>-2</sup>.
(4) Any difference between sum and Total is due to rounding.

Table 9: The Five Largest Wet NO3 Deposition Events at Kejimkujik, N.S. During 1993

		Preci	pitation	<u></u>		Wet NO3	Deposition
Even #	t Date M/D	Туре (1)	Amount (mm)	Upwind Sectors	[NO <sub>3</sub> <sup>-</sup> -N] mg·l <sup>-1</sup>	Amount mg·m <sup>-2</sup>	% of Total (3)
				(2)			
1	06/28	R	12.4	IV	0.59	32.4	3.6%
2	11/12	R	7.2	III	0.92	29.3	3.3%
3	06/18	R	2.5	IV	2.39	26.5	3.0%
4	01/29	S	8.0	IV	0.69	24.5	2.7%
5	10/30	R	23.8	IV,I	0.22	23.2	2.6%
				Total:		135.9	15.2% (4)
NOTE	S:						
(1)	Precipita	ation	Type: R	R = Rain S = Snow			
(2)	Upwind se	ectors	are def	ined in 7	Table 1.		
(3)	Total mea	asured	l deposit	ion was 8	391.5 mg	m <sup>-2</sup> .	
(4)	Any diff	erence	betweer.	sum and	Total is	due to a	rounding.

Based on back-trajectory information, the source regions that contributed to the large deposition amounts were mostly located at latitudes south of Kejimkujik, and mainly in Sector IV (Figure 2). Trajectory plots for the top ranking episode show that the air trajectories for this episode at the 925 and 850 mb levels in the lower atmosphere came from the eastern U.S.A.

The episodic character of the precipitation events themselves can be investigated (Table 10) and compared to the pollutant Eleven out of 205 precipitation days (about 5.4%) episodicity. accounted for approximately 30% of the total precipitation amount of 1445.5 mm. Thus, precipitation events were even more episodic hydrogen ion, in nature than excess sulphate and nitrate This was somewhat in contrast to 1992 at depositions in 1993. Kejimkujik, when precipitation events were not quite as episodic as either excess sulphate or nitrate depositions, but slightly more episodic than hydrogen ion deposition.

A previous study (OECD, 1979) found that the episodic nature of sulphate deposition was relatively independent of precipitation episodicity, indicating that the most pronounced pollution episodes are associated with special meteorological situations involving the long range transport of air pollutants. These findings are supported for Kejimkujik by the fact that only three of the 11 precipitation events which comprised the top 30% of total precipitation amount in 1993 were also pollutant episodes.

 Event #	Date M/D	Type (1)	Amount (mm)	% of Tota (2)	1	
1 2 3 4 5	10/27 11/05 12/05 03/13 01/27	R R R M R	66.8 49.0 46.6 43.6 37.6	4.6% 3.4% 3.2% 3.0% 2.6%		
 Tota	1:		243.6	16.9% (:	3)	

Table	10:	The Five Largest Precipitation Events
		at Kejimkujik, N.S. During 1993

NOTES:

(1) Precipitation Type: R = Rain

M = Mixed Rain and Snow

(2) Total measured precipitation was 1445.5 mm.

(3) Small difference between sum and Total is due to roundoff.

## 3.6 Trends over Period of Record

Figure 9 shows the average annual pH of precipitation received at Kejimkujik from 1980 to 1993. The least acidic of the fourteen



years was 1983 (average pH of 4.71) and the most acidic year was 1985 (average pH of 4.40). It is difficult to identify any trends over this time period. The variability in pH within each year, as well as from year to year, is shown in Appendix C.

Deposition values, rather than pH, present a clearer picture of the cumulative effect on the area, since it is the total deposition of acid from precipitation that causes the long term damage to the environment (MOI, 1983). Appendix C illustrates that the largest annual wet hydrogen ion deposition from 1980 to 1993  $(0.53 \text{ kg}\cdot\text{ha}^{-1})$  occurred in 1986. The deposition in recent years has not fallen to the low levels observed in 1983  $(0.26 \text{ kg}\cdot\text{ha}^{-1})$  and 1984  $(0.29 \text{ kg}\cdot\text{ha}^{-1})$ .

Annual wet deposition amounts for excess sulphate and nitrate are shown in Figure 10 and Appendix C. For excess sulphate, the annual wet deposition from 1980-1993 varied from a high in 1981 of 20.2 kg ha<sup>-1</sup>, to a low in 1993 of 9.6 kg ha<sup>-1</sup>; thus, 1993 received the lowest amount of excess sulphate deposition for the 14-year period of record. For nitrate, the annual wet deposition varied from a high in 1989 of 14.4 kg ha<sup>-1</sup> to a low in 1983 of 5.4 kg ha<sup>-1</sup>. (For Figure 10 and Appendix C, data for previous years were obtained as follows: 1980-1983 data from A.E.S. (n.d.); 1984-1987 data from Vet et al. (1986; 1988a; 1988b; 1989); and 1988-1992 data from A.E.S. (1989; 1990; 1991; 1992; 1993a).)

Given the variability of the annual deposition data, it is difficult to decipher the long-term trends. However, a comparison of the average excess sulphate and nitrate depositions for the first five years of the period 1980 to 1993 to the average depositions for the last five years of the period showed that the average annual excess sulphate deposition decreased by almost 16% from the first five years to the last five years of the period, whereas the average annual nitrate deposition increased by over 31%. An examination of yearly precipitation totals for the period showed that there was little change (a 4% increase) from the first five years to the last five years.

The averaqe precipitation-weighted excess sulphate concentration for the five years 1980 to 1984 was 1.16 mg<sup>-1</sup>, whereas the average concentration for the five year period 1989 to 1993 was 0.94 mg·l<sup>-1</sup>, representing a reduction of about 19%. Average precipitation-weighted nitrate concentration for the five 0.14  $mg^{-1}$ , whereas the average 1980 years to 1984 was concentration for the five year period 1989 to 1993 was 0.18 mg  $1^{-1}$ , representing an increase of about 26%.

evidence Visual for decrease а in excess sulphate concentration at Kejimkujik is shown in Figure 11, where the annual average precipitation-weighted excess sulphate concentration is plotted for the 14-year period 1980-1993, and a spline curve is fitted to the data. (A spline curve is a curved line through every point in the plot such that the curve is smooth everywhere (Wilkinson, 1990).) The plot for nitrate concentration (Figure 12) shows evidence for a decrease in the early 1980's, then an increase to reach a maximum during the second half of the decade, exhibiting a trend similar to hydrogen ion concentration. Statistical





Figure 11 Annual Excess Sulphate Concentration in Precipitation at Kejimkujik, N.S. 1980-1993

Year



Figure 12 Annual NO3-N Concentration in Precipitation at Kejimkujik, N.S. 1980-1993

analyses of the time variation of daily precipitation-weighted concentrations and depositions of excess sulphate, nitrate and hydrogen ion for 1980 to 1992 revealed statistically significant long-term trends at Kejimkujik (Sirois, 1993), similar to the trends noted here.

The relative importance of excess sulphate versus nitrate deposition at Kejimkujik has decreased in recent years, as indicated by the difference between the ratio of sulphate to nitrate deposition for the period 1980 to 1984 (1.89) and 1989 to 1993 (1.20). The largest annual ratio (2.00) occurred in 1981 and the smallest (1.04) occurred in 1993. Based on ion equivalents, excess sulphate contributed 2.5 times as much as nitrate to precipitation acidity in 1981, but only 1.4 times as much in 1993. This change at Kejimkujik may be the result of a decrease in the ratio of Eastern North American SO<sub>2</sub> to NO<sub>x</sub> emissions during the period due to a much larger reduction in anthropogenic SO<sub>2</sub> emissions than in NO<sub>x</sub> emissions (Sirois and Summers, 1989; RMCC, 1990).

### 4. Conclusions

The average annual precipitation-weighted pH for 1993 at Kejimkujik National Park in southwestern Nova Scotia was 4.66, with the most acidic seasonal average in summer and the least acidic in the winter. The most acidic event of the year (pH of 3.13) was 204 times more acidic than the least acidic event (pH of 5.44). About 17% of the events were highly acidic (pH  $\leq$  4.0). In comparison with the previous year, Kejimkujik experienced only a 2% increase in H<sup>+</sup> deposition despite a 26% increase in precipitation, primarily because of a 19% decrease in H<sup>+</sup> concentration in 1993.

Analyses indicated that a relationship existed between the acidity of precipitation at Kejimkujik and the levels of pollutant emissions from upwind sources. Precipitation events with pHs in the highly acidic range (pH  $\leq$  4.0) generally had air trajectories that crossed the more heavily industrialized sectors of the United States and Canada. Conversely, precipitation events with only slightly acidic pH values (pH > 4.7) predominantly had air trajectories from the Atlantic Ocean.

Wet deposition of excess sulphate and nitrate during 1993 at Kejimkujik was 9.6 and 9.2 kg ha<sup>-1</sup>, respectively; less than the 14year means of 14.6 and 9.9 kg ha<sup>-1</sup>, respectively. Average annual precipitation-weighted concentrations were 0.65 mg 1<sup>-1</sup> for excess sulphate and 0.14 mg 1<sup>-1</sup> for nitrate (expressed as nitrogen). A significant fraction of the annual deposition (e.g. 30%) was contributed by a small percentage of the precipitation days (e.g. 6% to 8% depending on the chemical). The largest daily deposition occurred on June 18, when 0.4 kg ha<sup>-1</sup> of excess sulphate (over 4% of the annual total) and 0.06 kg ha<sup>-1</sup> of nitrate (3% of the annual total) were deposited. The pollutant source region for this large deposition episode was the upwind sector with the largest emissions (eastern U.S.A. and southern Ontario).

The least acidic year from 1980 to 1993 was 1983 (average pH of 4.71) and the most acidic year was 1985 (average pH of 4.40). The annual wet excess sulphate deposition varied from a high in 1981 of 20.2 kg ha<sup>-1</sup> to a low in 1993 of 9.6 kg ha<sup>-1</sup>; thus, 1993 received the lowest amount of excess sulphate deposition for the 14-year period of record. For nitrate, the annual wet deposition varied from a high in 1989 of 14.4 kg ha<sup>-1</sup> to a low in 1983 of 5.4 kg ha<sup>-1</sup>. The large variability in the annual excess sulphate deposition data tends to obscure the decreasing trend evident in the concentration data.

The relative importance of excess sulphate versus nitrate deposition at Kejimkujik has decreased in recent years, likely as a result of a much larger reduction in anthropogenic SO<sub>2</sub> emissions than in nitrogen oxide emissions in Eastern North America.

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## APPENDIX A

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Canadian Air and Precipitation Monitoring Network (CAPMoN) Summary Statistics

Kejimkujik, N.S.

1993

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				CANADIAN	AIR AND P	RECIPITAT	TINOH NOIT	oring net cs	WORK (CAI	(NOH)				-	4NOV94
STATION NAME: SUMMARY TYPE: SUMMARY PERIOD: START: END:	(EJIMKUJIK (EAR 4LL/1993 31JAN93 31DEC93										TOT # TERA NON # TOTA	AL PRECIP CE PRECIP RACE PREC L DEPTH -	DAYS : 2 DAYS : DAYS : 2 STND : 1	18 6 12 48.37 cm	
	GAUGE	SAMPLE	COLL		CON	CENTRATIC	N IN MILL	IGRAMS PE	R LITRE						
	E	(NM)	(\$)	Н.	#	\$04=	XS04=	N03-	년	+44N	NA+	CA++	WG++	¥	%IBAL
# VALID SAMPLES	218	215	208	178	178	205	205	205	205	193	187	188	189	184	166
<pre>% VALID SAMPLES % TOTAL PRECIP-VALID % COLL EFF - VALID</pre>				97 90 90	90 84 90	66 16 16	16 16	16 16	16 66	16 16	8668	88 6 88	98 90 90	68 16 16 16 16	86 96 96
ARITHNETIC MEAN	7.0	6.1	84.2	4.50	0.0637	2.78	2.22	2.44	3.85	0.224	1.70	0.16	0.25	0.08	-6.37
ARITHMETIC S.D.	10.9	9.9	34.1	0.51	0.0974	3.96	3.63	4.25	9.40	0.344	5.06	0.33	0.67	0.13	7.31
GEOMETRIC MEAN	2.1	2.0	84.0	4.47	0.0320	1.27	0.93	0.89	0.79	0.066	0.33	0.06	0.06	0.03	
GEOMETRIC S.D. DISTRIBUTION PROBABILITY	1.7	1.7	0.4	0.12	1.1773	1.33	1.39	1.58	1.77	1.842	1.81	1.33	1.59	1.33	
- Normal - Log-Normal	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	<0.01* 0.03*	<0.01* 0.45	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	<0.01* <0.01*	
HINING	T	0.0	0.0	3.13	0.0036	0.05	0.00	<0.04	0.01	<0.001	0.01	<0.01	<0.01	<0.01	-33.30
5TH PERCENTILE	0.2	0.0	0.0	3.71	0.0049	0.11	0.09	<0.44	0.06	0.001	0.03	0.01	<0.01	<0.01	-19.70
10TH PERCENTILE	0.2	0.1	42.9	3.84	0.0063	0.21	0.14	0.09	0.10	0.005	0.04	0.01	0.01	<0.01	-15.70
25TH PERCENTILE	0.4 -	0.3	62.8 08 9	4.14 A 50	0.0117	0.50	0.36 0 av	0.27	0.21 0.67	0.021	0.08	0.02	0.02	0.01	0 <b>4</b> 0 
75TH PERCENTILE	7.2	6.3	104.0	4.93	0.0724	3.18	2.37	3.10	2.41	0.270	1.13	0.15	0.16	0.0	-2.00
90TH PERCENTILE	23.8	19.7	112.5	5.20	0.1445	6.61	6.15	5.71	9.50	0.742	3.80	0.40	0.54	0.20	0.00
95TH PERCENTILE MAXIMUM	32.2 66.8	27.2 66.0	123.1 179.2	5.31 5. <b>44</b>	0.1950 0.7413	8.61 27.92	8.01 27.78	8.28 40.28	19.64 86.00	0.966 2.122	7.35 58.14	0.55 2.96	1.10	0.29 1.00	0.90 22.00
dw nean-stind (ng/l')				4.66+	0.0218	0.82	0.65	0.62	1.26	0.076	0.67	0.05	0.09	0.04	
DEPN - STND (KG/HA)					0.3234	12.17	9.64	9.20	18.69	1.128	9.94	0.74	1.34	0.59	
NOTE : * - DATA NOT CI	NSISTENT V	VITH STATE	D DISTRIB	UTION AT	95% SIGNI	FICANCE L	EVEL.								
- NOT APPLICI + - PW MEAN PH	(IN PH UN)	r determin Its) calcu	ED. LATED FRO	H PH MEAN	CONCENTR	ATION OF	Ht.								
T - TRACE.		W NOT CAL		UPN TOTAT	DDECTD_U	01 / UI IV									
AAA - SITE NOT OF	ERATIONAL	FOR ALL C	E SUMMARY	PERIOD	1-11-11L	0/ < ATTW	•								

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## APPENDIX B

# Precipitation Chemistry Data Set

Kejimkujik, N.S.

1993

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DATE	PCP	PCP	PH	S	ECTOR	H+ DEPN	XSO4	XSO4	NO3-N	NO3-N
	ТҮР	AMT (mm)				(mg/m**2)	CONC (ma/l)	(ma/m**2)	(mail)	(maim**2)
01/01/93	<u> </u>	(1111)	4 45			0.0284	0.94	0.75	0.37	0.30
01/02/93	š	1.4	4.67	3	2	0.0300	0.72	1.01	0.17	0.24
01/03/93	Ř	0.8	4.19	•	-	0.0517	2.54	2.03	0.30	0.24
01/04/93	R	0.4	4.00			0.0400	3.88	1.55	0.64	0.26
01/05/93	R	25.0	5.26	4		0.1375	0.15	3.75	0.03	0.75
01/06/93	S	0.2	4.16			0.0138	3.09	0.62	0.27	0.05
01/08/93	S	0.2	-9.00			-9.0000	4.22	0.84	1.08	0.22
01/13/93	S	11.8	5.25	2		0.0661	0.09	1.06	0.04	0.47
01/15/93	S	1.0	3.84	4		0.1445	2.68	2.68	1.29	1.29
01/16/93	S	0.4	-9.00			-9.0000	7.05	2.82	0.76	0.30
01/17/93	S	1.2	3.86	3		0.1656	3.78	4.54	1.41	1.69
01/18/93	S	1.8	4.32	3		0.0862	0.56	1.01	0.56	1.01
01/19/93	S	2.6	4.64	3		0.0595	0.28	0.73	0.20	0.52
01/20/93	S	1.4	4.62	3		0.0336	0.16	0.22	0.22	0.31
01/22/93	R	27.4	5.26	1		0.1507	0.16	4.38	-0.01	0.14
01/23/93	R	3.4	5.26	4		0.0187	0.11	0.37	0.02	0.07
01/24/93	R	5.8	4.62	4	1	0.1392	0.98	5.68	0.12	0.70
01/27/93	S	37.6	5.44	4	1	0.1354	0.02	0.75	-0.01	0.19
01/28/93	S	0.6	4.07			0.0511	0.95	0.57	1.26	0.76
01/29/93	S	8.0	4.15	4		0.5664	1.59	12.72	0.69	5.52
01/30/93	S	4.4	4.72	3		0.0840	0.44	1.94	0.15	0.66
01/31/93	S	2.6	4.22	4	1	0.1568	0.36	0.94	0.69	1.79
02/01/93	S	0.4	4.15			0.0283	0.26	0.10	0.83	0.33
02/03/93	S	1.0	4.12	3		0.0759	0.87	0.87	0.96	0.96
02/04/93	S	0.4	4.21			0.0247	1.12	0.45	0.71	0.28
02/05/93	S	4.4	4.11	3		0.3414	0.91	4.00	0.97	4.27
02/07/93	S	0.2	-9.00			~9.0000	1.17	0.23	0.31	0.06
02/08/93	S	2.8	4.25	4		0.1574	1.06	2.97	0.46	1.29
02/09/93	S	0.4	-9.00			-9.0000	1.29	0.52	0.48	0.19
02/10/93	S	0.4	-9.00			-9.0000	9.10	3.64	3.60	1.44
02/12/93	М	17.0	5.25	1		0.0952	0.12	2.04	0.02	0.34
02/13/93	R	11.8	5.20	1		0.0743	0.15	1.77	0.04	0.47
02/14/93	S	0.4	-9.00			-9.0000	6.15	2.46	1.04	0.42
02/16/93	M	34.8	5.04	1		0.3167	0.37	12.88	0.02	0.70
02/17/93	M	2.0	4.34	3		0.0914	1.73	3.46	0.33	0.66
02/18/93	S	0.4	-9.00			-9.0000	1.02	0.41	0.81	0.32
02/19/93	S	4.1	-9.00	3	2	-9.0000	0.28	1.15	0.71	2.91
02/20/93	S	0.6	4.24			0.0345	0.31	0.19	0.70	0.42
02/21/93	S	8.2	5.19	4	1	0.0533	0.07	0.57	0.04	0.33
02/22/93	M	3.4	4.67	1		0.0728	0.96	3.26	0.11	0.37
02/23/93	S	1.0	3.71	4		0.1950	7.52	7.52	1.57	1.57
02/24/93	S	0.4	3.73			0.0745	3.20	1.28	2.26	0.90
02/25/93	S	0.2	4.17			0.0135	0.88	0.18	0.76	0.15
02/28/93	S	18.2	5.32	2	1	0.0874	0.08	1.46	-0.01	0.09
03/01/93	5	0.4	4.97			0.0043	0.41	0.16	-0.05	0.01
03/05/93	5	28.6	5.23	2	1	0.1687	0.10	2.86	0.02	0.57
03/07/02	5	7.6	5.10	1		0.0600	0.18	1.37	0.03	0.23
0001193	3	U.4	4.24			0.0230	1.55	0.62	0.43	0.17

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DATE	PCP	PCP	PH	S	ECTOR	H+ DEPN	XSO4	XSO4	NO3-N	NO3-N
	116	(mm)				(mg/m**2)	CONC (mg/l)	UEPN (ma/m**2)	CONC	DEPN
03/09/93	S	0.8	4.23			0.0471	1 18		(ing/i)	$\frac{(\text{ing/in}^2)}{0.43}$
03/10/93	ŝ	3.0	4.56	4	3	0.0825	0.70	2 10	0.01	0.41
03/11/93	Š	36.6	5.14	1	•	0 2635	0.10	6 95	0.02	0.42
03/12/93	ŝ	0.2	-9.00	•		-9.0000	1 94	0.39	0.39	0.75
03/13/93	M	43.6	5.11	1		0 3401	0 17	7 41	0.00	1 74
03/14/93	S	5.0	4 47	3	2	0 1695	1.32	6 60	0.04	1.74
03/15/93	ŝ	0.2	-9.00	-	-	~9 0000	1.62	0.34	0.20	0.06
03/17/93	M	27.4	5.05	4	1	0 2439	0.22	6.03	0.20	1 10
03/18/93	S	6.6	5.31	3	2	0.0323	0.05	0.33	-0.04	0.03
03/21/93	Ř	4.4	4 83	4	1	0.0651	0.59	2 60	0.05	0.00
03/24/93	R	1.2	4 73	1	•	0.0223	0.56	0.67	0.05	0.06
03/25/93	R	12	4 34	1		0.0548	1 74	2 09	0.11	0.13
03/29/93	R	4.4	4.31	1		0.2156	1.43	6.29	0.19	0.84
04/01/93	M	30.2	4.98	1		0.3171	0.24	7.25	0.05	1.51
04/02/93	R	0.8	3.98			0.0838	3.86	3.09	0.40	0.32
04/03/93	R	3.0	4.32	2	1	0.1437	1.25	3.75	0.26	0.78
04/10/93	R	2.4	4.75	1	•	0.0427	0.74	1.78	0.16	0.38
04/11/93	R	6.4	4.82	1		0.0966	0.66	4.22	0.09	0.58
04/12/93	R	1.0	4.69	1		0.0204	0.53	0.53	0.08	0.08
04/13/93	M	2.4	4.95	2		0.0269	0.13	0.31	0.10	0.24
04/14/93	R	0.8	5.12	-		0.0061	0.12	0.10	0.03	0.02
04/16/93	R	0.2	3.19			0.1291	23.41	4.68	4.25	0.85
04/17/93	R	4.6	4.71	4	1	0.0897	0.73	3.36	0.11	0.51
04/21/93	R	23.2	5.14	1		0.1670	0.21	4.87	0.03	0.70
04/22/93	R	12.0	5.05	1		0.1068	0.35	4.20	0.04	0.48
04/23/93	R	1.8	4.09	2		0.1463	1.60	2.88	0.70	1.26
04/24/93	R	0.4	4.34			0.0183	1.60	0.64	0.18	0.07
04/26/93	M	5.8	4.16	4	1	0.4014	3.02	17.52	0.38	2.20
04/27/93	M	26.8	4.99	1		0.2734	0.38	10.18	-0.01	0.13
04/29/93	R	0.2	-9.00			-9.0000	0.92	0.18	-0.10	0.01
04/30/93	R	13.0	5.18			0.0858	0.14	1.82	-0.01	0.07
05/01/93	R	4.0	5.14			0.0288	0.10	0.40	0.03	0.12
05/05/93	R	0.6	4.57			0.0161	1.00	0.60	0.17	0.10
05/06/93	R	5.6	4.17	4		0.3786	1.93	10.81	0.45	2.52
05/07/93	R	-0.2	4.53			0.0030	1.26	0.13	0.11	0.01
05/10/93	R	1.4	4.41	3		0.0545	1.54	2.16	0.15	0.21
05/11/93	R	6.0	4.27	2		0.3222	2.04	12.24	0.23	1.38
05/12/93	R	0.6	4.14			0.0434	3.77	2.26	1.08	0.65
05/13/93	R	0.6	3.82			0.0908	7.17	4.30	1.51	0.91
05/16/93	R	3.8	3.92	4		0.4568	6.28	23.86	0.97	3.69
05/17/93	R	0.2	-9.00			-9.0000	2.09	0.42	0.26	0.05
05/18/93	R	10.0	4.53	4		0.2950	0.71	7.10	0.30	3.00
05/19/93	R	12.8	4.63	1		0.2995	1.04	13.31	0.16	2.05
05/20/93	R	11.2	4.79	4	1	0.1814	0.58	6.50	0.09	1.01
05/21/93	R	12.5	5.11	1		0.0975	0.28	3.50	0.04	0.50
05/22/93	R	2.6	4.65	4		0.0582	0.73	1.90	0.16	0.42
05/23/93	R	1.8	4.65	2		0.0403	0.95	1.71	0.14	0.25
05/24/93	R	2.4	3.75	4		0.4267	7.50	18.00	1.09	2.62

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DATE	PCP	PCP	PH	S	ECTOR	H+ DEPN	XSO4	XSO4	NO3-N	NO3-N
	TYP	AMT				(mg/m**2)	CONC	DEPN	CONC	DEPN
		(mm)					(mg/l)	(mg/m**2)	(mg/l)	(mg/m**2)
05/25/93	R	0.4	3.82			0.0606	6.57	2.63	0.88	0.35
05/29/93	R	5.0	4.11	3	2	0.3880	2.99	14.95	0.52	2.60
05/30/93	R	1.2	4.61	2		0.0294	0.73	0.88	0.12	0.14
05/31/93	R	1.4	4.37	1		0.0598	1.28	1.79	0.28	0.39
06/01/93	R	12.4	4.99	4	1	0.1265	0.26	3.22	0.05	0.62
06/06/93	R	32.2	4.90	1		0.4057	0.37	11.91	0.06	1.93
06/07/93	R	1.0	4.33	2	1	0.0468	1.50	1.50	0.19	0.19
06/09/93	R	0.4	3.68			0.0836	5.82	2.33	1.14	0.46
06/10/93	R	0.2	4.05			0.0178	5.38	1.08	0.89	0.18
06/15/93	R	1.0	4.62	4	1	0.0240	0.66	0.66	0.16	0.16
06/16/93	R	0.2	-9.00			-9.0000	8.01	1.60	2.69	0.54
06/18/93	R	2.5	3.34	4		1.1428	16.35	40.88	2.39	5.98
06/19/93	R	2.3	3.87	4		0.3103	3.67	8.44	0.76	1.75
06/21/93	R	4.8	3.78	4	1	0.7968	5.67	27.22	0.75	3.60
06/22/93	R	0.6	4.91			0.0074	0.59	0.35	-0.01	0.00
06/23/93	R	4.0	5.14	2		0.0288	0.27	1.08	-0.01	0.02
06/27/93	R	20.0	5.34	4	1	0.0920	0.16	3.20	0.03	0.60
06/28/93	R	12.4	4.03	4		1.1569	2.54	31.50	0.59	7.32
06/29/93	R	0.2	-9.00			-9.0000	4.11	0.82	1.00	0.20
07/03/93	R	16.3	4.58	1		0.4287	0.85	13.86	0.11	1.79
07/08/93	R	0.4	3.96			0.0438	4.18	1.67	0.43	0.17
07/10/93	R	2.4	4.07	3		· 0.2042	3.17	7.61	0.37	0.89
07/12/93	R	0.2	-9.00			-9.0000	14.02	2.80	1.87	0.37
07/14/93	R	1.4	4.12	3		0.1063	2.20	3.08	0.50	0.70
07/15/93	R	0.8	3.58			0.2104	8.36	6.69	0.90	0.72
07/16/93	R	1.4	4.62	2		0.0336	0.52	0.73	0.05	0.07
07/17/93	R	-0.2	-9.00			-9.0000	3.09	0.31	0.11	0.01
07/20/93	R	6.2	5.04	1		0.0564	0.25	1.55	0.04	0.25
07/21/93	R	0.4	-9.00			-9.0000	2.35	0.94	0.73	0.29
07/22/93	R	2.8	4.54	4	3	0.0806	0.56	1.57	0.21	0.59
07/27/93	R	1.8	4.21	1		0.1111	1.60	2.88	0.30	0.54
07/28/93	R	12.2	4.59	4	1	0.3135	0.76	9.27	0.13	1.59
07/29/93	R	2.2	3.90	4	1	0.2770	3.52	7.74	0.64	1.41
08/01/93	R	8.2	4.48	1		0.2714	0.46	3.77	0.26	2.13
08/02/93	R	20.8	4.61	4		0.5096	0.59	12.27	0.16	3.33
08/04/93	R	3.8	4.17	4	1	0.2569	1.83	6.95	0.33	1.25
08/12/93	R	0.2	-9.00			-9.0000	0.86	0.17	-0.05	0.01
08/13/93	R	0.2	-9.00			-9.0000	6.29	1.26	1.09	0.22
08/19/93	R	0.2	4.22			0.0121	1.86	0.37	0.34	0.07
08/20/93	R	11.6	4.11	4		0.9002	2.37	27.49	0.33	3.83
08/24/93	R	0.6	3.35			0.2680	9.90	5. <del>9</del> 4	3.57	2.14
09/01/93	R	1.8	3.74	4	3	0.3276	6.09	10.96	0.64	1.15
09/03/93	R	0.8	3.87			0.1079	2.88	2.30	1.09	0.87
09/04/93	R	5.0	4.26	4		0.2750	1.46	7.30	0.22	1.10
09/07/93	R	0.2	-9.00			-9.0000	5.12	1.02	0.70	0.14
09/08/93	R	2.8	3.61	4		0.6874	7.42	20.78	1.33	3.72
09/09/93	R	0.2	-9.00			-9.0000	17.47	3.49	2.32	0.46
09/10/93	R	5.0	4.46	4		0.1735	0.83	4.15	0.16	0.80

DATE	PCP	PCP	PH	S	ECTOR	H+ DEPN	XSO4	XSO4	NO3-N	NO3-N
	TYP	AMT				(mg/m**2)	CONC	DEPN	CONC	DEPN
		(mm)					(mg/i)	(mg/m**2)	(mg/l)	(mg/m**2)
09/15/93	R	0.2	4.23			0.0118	2.41	0.48	0.46	0.09
09/16/93	R	0.2	-9.00			-9.0000	0.72	0.14	0.30	0.06
09/17/93	R	27.2	5.30	1		0.1360	0.11	2.99	0.01	0.27
09/18/93	R	1.0	4.49	4	1	0.0324	0.58	0.58	0.26	0.26
09/23/93	R	0.8	3.44			0.2905	8.49	6.79	2.70	2.16
09/26/93	R	14.8	4.52	4	1	0.4470	0.75	11.10	0.17	2.52
09/27/93	R	33.2	4.74	4		0.6042	0.68	22.58	0.11	3.65
09/28/93	R	0.4	4.46			0.0139	1.77	0.71	0.26	0.10
10/02/93	R	1.2	4.43	1		0.0446	0.86	1.03	0.30	0.36
10/03/93	R	6.6	4.28	4	1	0.3465	1.71	11.29	0.31	2.05
10/04/93	R	1.6	3.95	4		0.1795	3.55	5.68	1.19	1.90
10/05/93	R	2.4	4.29	3		0.1231	1.62	3.89	0.39	0.94
10/06/93	R	0.2	-9.00			-9.0000	2.00	0.40	0.14	0.03
10/07/93	R	0.2	3.13			0.1483	27.78	5.56	9.10	1.82
10/08/93	R	0.2	-9.00			-9.0000	12.40	2.48	5.51	1.10
10/09/93	R	3.4	4.27	4	1	0.1826	1.63	5.54	0.34	1.16
10/12/93	R	5.0	4.78	1		0.0830	0.39	1.95	0.12	0.60
10/17/93	R	29.4	5.01	1		0.2881	0.35	10.29	0.03	0.88
10/18/93	R	8.6	5.32	4	1	0.0413	0.09	0.77	0.01	0.09
10/21/93	R	17.0	4.54	1		0.4896	0.93	15.81	0.17	2.89
10/23/93	R	1.8	4.95	3		0.0202	0.38	0.68	0.03	0.05
10/27/93	R	66.8	5.35	1		0.3006	0.08	5.34	0.01	0.67
10/28/93	R	0.4	4.21			0.0247	1.63	0.65	0.59	0.24
10/30/93	R	23.8	4.62	4	1	0.5712	0.67	15.95	0.22	5.24
10/31/93	R	1.6	4.59	1		0.0411	1.10	1.76	0.12	0.19
11/01/93	R	4.6	4.21	4	1	0.2838	1.16	5.34	0.52	2.39
11/02/93	R	0.4	-9.00			-9.0000	0.84	0.34	80.0	0.03
11/03/93	R	4.0	4.77	1		0.0680	0.35	1.40	0.12	0.48
11/05/93	R	49.0	4.93	4	1	0.5733	0.34	16.66	0.07	3.43
11/06/93	R	0.2	3.74	_		0.0364	6.39	1.28	1.32	0.26
11/12/93	R	7.2	4.07	3		0.6127	2.15	15.48	0.92	6.62
11/13/93	R	4.4	4.23	4	1	0.2592	1.50	6.60	0.54	2.38
11/14/93	R	1.2	4.00	4	3	0.1200	3.11	3.73	0.89	1.07
11/15/93	R	3.8	4.07	4		0.3234	2.82	10.72	0.47	1.79
11/17/93	R	20.0	4.94	4		0.2300	0.34	6.80	0.05	1.00
11/19/93	R	16.4	4.93	1		0.1919	0.37	6.07	0.06	0.98
11/20/93	S	1.8	4.14	4		0.1303	1.21	2.18	0.77	1.39
11/28/93	R	30.6	5.39	1		0.1255	0.37	11.32	0.02	0.61
12/02/93	R	0.4	4.41			0.0156	1.27	0.51	0.17	0.07
12/03/93	ĸ	1.2	5.00	4		0.0720	0.30	2.10	0.04	0.29
12/04/93	ĸ	4.0	4.51	4		0.1421	0.70	3.22	0.20	1.20
12/05/93	R	46.6	5.31	1		0.2283	0.10	4.00	0.01	0.47
12/06/93	S	0.2	3.90			0.0252	0.00	0.00	0.15	0.03
12/07/93	3	0.6	4.10			0.0397	2.14	1.28	4.49	0.10
12/08/93	ა ი	0,0	3.00			0.0020	1.80	1.14	1.40	0.09
12/10/93	л Р	U.Z	-9.00 5.25	4		-9.0000	0.00	0.07	1.41	0.20
12/13/33	л Р	۲.0 ۲۸	5.20		1	0.0001	0.14	1.00	0.02	0.24
12/14/93	R	4.4	J. 14	~	1	0.0317	U.17	0.75	0.03	0.13

DATE	PCP TYP	PCP AMT	PH	SECTOR		H+ DEPN (mg/m**2)	XSO4 CONC	XSO4 DEPN	NO3-N CONC	NO3-N DEPN		
		(mm)				· • /	(mg/l)	(mg/m**2)	(mg/l)	(mg/m**2)		
12/15/93	R	0.6	5.15			0.0043	0.23	0.14	0.04	0.02		
12/19/93	R	19.4	4.87	4	1	0.2619	0.39	7.57	0.06	1.16		
12/21/93	R	37.0	4.72	4	1	0.7067	0.51	18.87	0.11	4.07		
12/22/93	М	0.6	3.79			0.0973	4.36	2.62	1.39	0.83		
12/23/93	S	0.2	-9.00			-9.0000	0.44	0.09	1.87	0.37		
12/24/93	S	2.4	4.61	4		0.0588	0.02	0.05	0.28	0,67		
12/25/93	S	14.4	4.86	4	1	0.1987	0.35	5.04	0.06	0.86		
12/26/93	S	9.6	4.48	4	3	0.3178	0.57	5.47	0.33	3.17		
12/27/93	S	0.8	4.68			0.0167	0.17	0.14	0.16	0.13		
12/28/93	S	1.8	4.74	3		0.0328	0.07	0.13	0.16	0.29		
12/29/93	S	26.0	5.42	1		0.0988	0.04	1.04	0.02	0.52		
12/30/93	S	16.8	5.06	3	2	0.1462	0.11	1.85	0.08	1.34		
12/31/93	S	1.4	4.04	4	3	0.1277	0.59	0.83	0.90	1.26		
R = rain, S = snow, M = mixed rain and snow												

NOTE: See text (Section 2.2) for explanation of negative values.

## APPENDIX C

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Annual Precipitation-weighted pH, Annual H<sup>+</sup>, XSO<sub>4</sub><sup>=</sup> and NO<sub>3</sub><sup>-</sup> Depositions and Annual Precipitation Kejimkujik, N.S. 1980-1993

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Precipitation (mm)		1190.6	1625.2	1260.2	1344.1	1315.0	1298.7	1454.0	1238.2	1428.2	1304.8	1607.3	1436.8	1176.7	1484.2	1368.9
1a <sup>-1</sup> )	NO <sub>3</sub> -	10.0	10.1	7.8	5.4	8.2	12.1	11.6	9.3	10.1	14.4	10.7	10.8	9.4	9.2	6.9
Deposition (kg <sup>1</sup>	XSO4 <sup>=</sup>	18.0	20.2	14.6	10.6	14.7	17.0	17.0	12.3	14.0	16.7	13.8	14.8	10.8	9.6	14.6
	H <sup>+</sup>	0.43	0.41	0.35	0.26	0.29	0.51	0.53	0.33	0.37	0.47	0.38	0.38	0.32	0.32	0.38
ΡΗ	Maximum	5.60	7.40	7.30	6.00	6.50	5.48	5.39	5.57	5.63	5.50	5.82	5.50	5.47	5.44	
	Mean	4.45	4.60	4.56	4.71	4.66	4.40	4.44	4.58	4.59	4.44	4.63	4.58	4.57	4.66	4.56
	Minimum	3.60	3.60	3.40	3.60	3.15	3.49	3.31	3.55	3.41	3.36	3.40	3.52	<u>3.04</u>	3.13	
Year		1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	Average

NOTE: The maximum value in each column is typed in **bold**; the minimum value in each column is <u>underlined</u>. The mean pH for each year and the average mean pH at the bottom of column 3 are precipitation-weighted averages.

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Acid precipitation during 1993 at Keiimkujik. N.S. BEATTIE. BILLIE L

ЭС 985.5.M4 M34 95-2 С.2 NSHW 1607098I 1600390H