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Acid Precipitation during 1992 at Kejimkujik, N.S.

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Abstract

The average precipitation-weighted pH for 1992 at Kejimkujik National Park in southwestern Nova Scotia was 4.57, which is equal to the mean pH over the 13 years of record (1980-1992). The most acidic event of the year (pH of 3.04) was 269 times more acidic than the least acidic event (pH of 5.47). Highly acidic precipitation events generally had air trajectories that crossed the more heavily industrialized sectors of the United States and Canada.

Wet deposition of excess sulphate and nitrate during 1992 at Kejimkujik was 10.8 kg ha⁻¹ and 9.4 kg ha⁻¹, respectively, less than the 13-year means of 15.0 kg ha⁻¹ and 10.0 kg ha⁻¹, respectively. The largest daily deposition during 1992 occurred on June 15, when 0.6 kg ha⁻¹ of excess sulphate (almost 6% of the annual total) and 0.09 kg ha⁻¹ of nitrate (almost 5% of the annual total) were deposited.

A comparison of the average depositions for the first five years of the period of record to the average depositions for the most recent five years of the period showed that the average annual excess sulphate deposition decreased by over 10%, whereas the average annual nitrate deposition increased by over 33%. An examination of yearly precipitation totals for the period shows that there was little change (a 3% increase) from the first five years to the last five years.

Average precipitation-weighted concentrations for 1992 were $0.92 \text{ mg}\cdot l^{-1}$ for excess sulphate and $0.18 \text{ mg}\cdot l^{-1}$ 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^{-1}, whereas the average concentration for the five year period 1988 to 1992 was 1.01 mg l^{-1}, representing a reduction of about 13%. The concentration of nitrate has generally been increasing in recent years. Average precipitation-weighted nitrate concentration for the five years 1980 to 1984 was 0.14 mg l^{-1}, whereas the average concentration years 1980 to 2000 to 1984 was 0.14 mg l^{-1}, whereas the average concentration for the five year period 1988 to 1992 was 0.18 mg l^{-1}, representing an increase of about 29%.

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 from their sources, travel downwind and they show up in precipitation as sulphates (SO_4^{2}) , nitrates (NO_3^{2}) 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

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 al., 1990) is of marine biogenic origin, rather than of

anthropogenic origin. Further study is required to assess the biogenic component.

Recently, 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⁻¹·yr⁻¹ 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 began issuing, in December 1983, a weekly bulletin called the Acid Rain Report (see example in Appendix A). This bulletin lists 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 includes 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

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 1992 at Kejimkujik National Park; (2) to investigate the relationship between pollutant sources and resulting precipitation acidity at Kejimkujik; and (3) to compare the 1992 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 using an AES standard rain gauge and standard Nipher-shielded snow gauge, which are collocated with the precipitation chemistry collector. 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 $([H^+])$, excess (nonsea salt) sulphate concentration ([XSO4[±]]), 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 1992. The H^+ and $XSO_4^=$ concentrations were derived from measured parameters, as outlined in Vet et al. (1989): i.e., the concentration of H⁺ 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 procedure). This correction is recommended for sites located 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 21 out of the 209 precipitation days had insufficient precipitation amounts (less than 0.5 mm) for any chemical analysis. These entries were eliminated from the data set. The total precipitation for these 21 days was 4.5 mm, representing 0.4% of the total annual precipitation of 1177.0 mm. Of the remaining 188 precipitation days, one was rejected as invalid due to possible contamination (the collector was open when precipitation was not occurring), representing a precipitation amount of 18.6 mm. Two additional samples in the data set, though not rejected, had insufficient volume for pH measurements (precipitation total 0.4 mm). These missing pH values are designated by -9's in Appendix B.

The final precipitation chemistry data set for 1992 (Appendix B) thus contained 187 precipitation days, of which 185 had pH and $[H^+]$ values (representing 1153.5 mm of precipitation or 98% of the annual total), and the full 187 had XSO_4^{\pm} and $NO_3^{-}-N$ concentrations (representing 1153.9 mm of precipitation and also 98% of the annual total).

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 $mg \cdot m^{-2}$) by multiplying the chemical concentration (in $mg \cdot l^{-1}$) by the amount of precipitation (in mm) for each precipitation event. A water equivalent in millimetres was used for snow by assuming that 1 cm of snow was equivalent to 1 mm of water. (Units of kg \cdot ha^{-1} for wet deposition can be obtained from $mg \cdot m^{-2}$ by dividing by 100.)

To identify the pH class that made the dominant contribution to acidic deposition, the daily H^* depositions for the 185 days with valid pH measurements were summed for four pH classes (pH > 5.6; 4.7 < pH \leq 5.6; 4.0 < pH \leq 4.7; pH \leq 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 precipitationand then multiplied by weighted concentrations, 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^{-1} is 4.43 (the ratio of the molecular weight of NO_3^{-1} 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 m^{-2}) by multiplying the deposition (in mg m^{-2}) by the ion valence and then

dividing by the molecular weight.¹ Thus, the ratio of excess sulphate deposition to nitrate deposition was converted into its ion equivalent ratio by multiplying by 1.29.

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 132 precipitation days in 1992 (representing 1120.7 mm of precipitation or 95% of the annual total). All but three of the days without this information had small precipitation amounts (less than 1.0 mm). However, no trajectory information was available for June 24, June 25 and Aug. 29, when 6.6 mm, 3.8 mm and 1.0 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 185 days during 1992 with precipitation pH values, 53 did not have corresponding trajectory information and therefore, the upwind sectors were unknown. Of the remaining 132 days, the upwind source was unambiguous for 90 days (the trajectory to Kejimkujik crossed only one sector), but was ambiguous for the remaining 42 days (the trajectory crossed two sectors). In these 42 cases, the upwind sector with the highest sulphur oxide emissions (see Table 1) was assumed to be the major

¹ The valence for the sulphate ion is 2 and for the nitrate ion, one; the molecular weight for sulphate is 96.1 g mole⁻¹ and for nitrate, 62.0 g mole⁻¹.

Figure 2

Four Sectors Surrounding Kejimkujik, N.S.



source sector, unless the trajectory only touched the sector. Of the 187 days in 1992 with XSO_4^{\pm} and $NO_3^{-}N$ concentrations, 55 did not have corresponding trajectory information. As for pH, of the remaining 132 days, the upwind source was unambiguous for 90 days, and ambiguous for 42.

Sector	Description	Em: SO _x (ki:	issions NO _x 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	Anthropo	ogenic	Emissi	lons	of	SOx	and	NO _x
	from	Sectors	Surrou	inding	Kej	imku	ijik,	, N.	s.

Source of data: RMCC, 1990

3. Results and Discussion

3.1 Precipitation Event Characteristics

The considerable variability in H^+ , $XSO_4^=$ and NO_3^-N concentration and deposition amounts amongst precipitation events during the year is graphically illustrated in Figures 3-5, respectively. Two significant pH levels are indicated on Figure 3(b): moderately acidic (4.0 < pH \leq 4.7); and highly acidic (pH \leq 4.0).

An analysis of the distribution of pH in precipitation events (Figures 6a and 6b) shows that all of the precipitation events with valid pH data at Kejimkujik in 1992 were acidic (pH \leq 5.6) and that most were within the slightly acidic (4.7 < pH \leq 5.6) or moderately acidic (4.0 < pH \leq 4.7) ranges. 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 3.04 (May 25), which was 269 times more acidic than the least acidic event (pH 5.47 on February 8).







DAY



DAY





Table 2:	Summary of Precipitation pH Distribut	ion
	at Kejimkujik, N.S. During 1992	

Level of pH	pH Range	Number of Occurrences	Percentage
Normal (clean) Slightly Acidic Moderately Acidic Highly Acidic	pH > 5 4.7 < $pH \le 5$ 4.0 < $pH \le 4$ $pH \le 4$.6 0 .6 66 .7 88 .0 31	0% 36% 48% 17%
	Total:	185	101% (1)

NOTE:

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

Figures 7a and 7b show the distribution of daily wet hydrogen ion deposition for precipitation events with valid pH data, according to pH class, and show the relatively small contribution of the slightly acidic events and the dominant contribution of the moderately and highly acidic events, as summarized in Table 3.

Table 3: Summary of H⁺ Deposition by pH Class at Kejimkujik, N.S. During 1992

Level of pH	pH Range		Amor Depos	unt of sition	Percentage
Normal (clean)	pH >	5.6	0	mg·m ⁻²	0%
Slightly Acidic	4.7 < pH ≤	5.6	8	mg·m ⁻²	25%
Moderately Acidic	4.0 < pH ≤	4.7	11	mg·m ⁻²	36%
Highly Acidic	pH ≤	4.0	12	mg·m ⁻²	39%

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 8 to 10, respectively) graphically illustrate the usual relation between concentration and precipitation amount, and the effect on daily deposition amounts. (The largest deposition usually occurs for events with moderate concentration and moderate precipitation amount.)





Relation between H+ Concentration and Daily Precipitation Amount Kejimkujik, N.S. 1992 Figure 8



H+ Concentration (mg/l)

Relation between Sulphate Concentration and Daily Precipitation Amount 1992 Kejimkujik, N.S. Figure 9



Sulphate Concentration (mg/l)

Nitrate (NO3-N) Concentration (mg/l)



Figure 10

3.2 Concentration and Deposition by Precipitation Type

The three different types of precipitation that were recorded at Kejimkujik during 1992 (rain, snow, and mixed rain and snow) accounted for significantly different amounts of the total deposition of pollutants during the year.

The analysis for pH and H⁺ deposition by precipitation type is shown in Table 4. Rain events predominated at Kejimkujik and had the highest deposition (69% of the total). On average, during 1992, in contrast to 1991, rain was somewhat more acidic than snow (lower average pH).

Table 4: Average pH and H⁺ Deposition by Precipitation Type for Kejimkujik, N.S. During 1992

Precipitation				Average	Deposition	Percentage
Туре	Days	Amount	(mm)	pH	of H ⁺ (kg·ha ⁻¹)	Deposition
Rain	119	755.	1	4.54	0.219	69%
Snow	70	269.	3	4.59	0.070	22%
Mixed	20	152.	6	4.73	0.028	9%

The analysis for XSO_4^{\pm} and NO_3^{-} deposition is shown in Table 5. Rain events, not surprisingly, had the highest deposition (75% of annual deposition for excess sulphate and 64% of annual deposition for nitrate). Snow events had, on average, lower excess sulphate concentrations than rain events, but higher nitrate concentrations.

Table 5:	XSO4 ⁼ and NO3 ⁻ Precipitation-Weighted
	Average Concentrations (mg·l ⁻¹) and
	Deposition Amounts (kg ha ⁻¹) by Precipitation Type
	for Kejimkujik, N.S. During 1992
	(% of Annual Deposition in Brackets)

Precipitation Type Days Amount(mm)		pitation Amount(mm)	XSO4 ⁼ Conc. Deposition			Conc.	NO3 ⁻ Deposition		
Rain	119	755.1	1.07	8.1	(75%)	0.80	6.0 (64%) 2.7 (29%)		
Mixed	20	152.6	0.65	1.0	(9%)	0.44	0.7 (7%)		

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 6 shows the number of precipitation days, the total precipitation, the average precipitation-weighted pH and the deposition of H^* , 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 6) and the precipitation type depositions (Tables 4 and 5) may not sum to the annual depositions (Table 6).

Table 6: Average pH and Wet Deposition (kg^{-ha⁻¹}) of H⁺, XSO₄⁼ and NO₃⁻ by Season and for the Year at Kejimkujik, N.S. During 1992

	Pre	ecipitation	Average	We	Wet Deposition				
Period	Days	Amount (mm)	pH	H ⁺	XSO4=	NO ₃			
Winter	69	442.5	4.75	0.08	2.4	2.7			
Spring	45	197.2	4.30	0.10	4.0	2.6			
Summer	40	192.1	4.52	0.06	2.1	1.6			
Fall	55	345.2	4.63	0.08	2.4	2.4			
Annual	209	1177.0	4.57	0.32	10.8	9.4 (1)			

NOTE:

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

According to Table 6, spring had the lowest average pH and the largest deposition of H^{+} , while winter had the highest average pH. This is similar to 1991 (Beattie and O'Brien, 1992b), and to 1986 through 1989 (Beattie *et al.*, 1990; Hanley and Beattie, 1989; Allen and Beattie, 1988; and Pettipas and Beattie, 1987), though it is in contrast to 1990 (Beattie and O'Brien, 1992a). Spring also had the highest deposition of excess sulphate while the other three seasons had about an equal amount. For nitrate, the largest deposition occurred during the winter. The highest average precipitation-weighted concentrations (wet deposition divided by precipitation amount) for all three ions occurred during the spring.

Although the average annual pH at Kejimkujik in 1992 (4.57) was about the same as that for 1991 (4.58), deposition totals of H⁺ (0.32 kg ha⁻¹ compared to 0.38), XSO_4^{\pm} (10.8 kg ha⁻¹ compared to 14.8) and NO₃⁻ (9.4 kg ha⁻¹ compared to 10.8) were lower in 1992. Since concentrations were considerably closer than depositions would indicate, and in the case of [H⁺] and [NO₃⁻-N], were actually lower in 1991, this was a reflection of the smaller amount of precipitation in 1992 (1177.0 mm compared to 1436.8). (Data for 1991 were obtained from A.E.S. (1992).) To illustrate this point in more detail, the annual H⁺ deposition at Kejimkujik in 1992 (31.7 mg m⁻² or 0.32 kg ha⁻¹) represented a decrease of over 16% from the 1991 deposition of 37.9 mg m⁻² (A.E.S., 1992).

precipitation amount in 1992 (1177.0 mm) represented a decrease of 18% from the 1991 amount (1436.8 mm). Since the normal amount of precipitation at Kejimkujik National Park (based on 1966 - 1990) is 1396.9 mm (A.E.S., 1993), the precipitation in 1992 was 16% below normal.

Box plots showing the statistical distribution of pH, $XSQ_4^=$ concentration and deposition, and NO_3^--N concentration and deposition for each season and the year are shown in Figures 11 to 15. It can be seen that median values, as illustrated by these figures, sometimes show seasonal patterns which are somewhat different than for the mean values in Table 6, or are not quite as marked. Most notable is the case of nitrate concentrations (Figure 14), where the median was lowest in spring, but the spring mean was about double that of other seasons. This occurred because there were a number of low concentrations (giving a low median) and also some high concentrations (giving a higher mean).

Seasonal ratios of excess sulphate to nitrate deposition based on ion equivalents (Figure 16) 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 about twice as much to the acidity of the spring and summer deposition as nitrate; whereas, in the winter, nitrate contributed almost as much as excess sulphate. Similar results were found in earlier studies: for the period 1979-1987 (Sirois and Summers, 1989), for 1990 and 1991 (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 pie diagrams in Figures 17 to 19. These pies illustrate the air mass back-trajectory information for 1992 (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 diagram represents an ion concentration 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 and percentage of precipitation events (with the number 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 12 Seasonal and Annual Box Plots of XSO4 Concentration in Precipitation Kejimkujik, N.S. 1992









Seasonal and Annual Box Plots of Wet NO3-N Deposition Kejimkujik, N.S. 1992 Figure 15

.











Figure 17 illustrates the relationship between pH of precipitation at Kejimkujik and the levels of pollutant emissions from upwind sectors. For example, precipitation events at Kejimkujik with pH in the highly acidic range (pH \leq 4.0) generally had air mass trajectories that crossed the more heavily industrialized regions in the United States and Canada (Sector IV). This sector was the major source sector for 86% (12 out of 14) of the events with known source regions. Precipitation events with less acidic levels show a reduced influence from Sector IV. For instance, the trajectories to Kejimkujik had Sector IV (southern Ontario and the U.S.) as the major source sector in 56% (34 out of 61) of the events with known source regions that had pH in the moderately acidic range $(4.0 < pH \le 4.7)$. Precipitation events with only slightly acidic pH values (pH > 4.7) predominantly had the Atlantic Ocean (Sector I) as the major source sector. Moreover, only 26% (15 out of 57) of the events with known sectors that had precipitation pH in the slightly acidic range had Sector IV as the major source sector.

For Figures 18 and 19, ranges of XSO_4^{\pm} and $NO_3^{-}-N$ concentrations were determined by choosing the 25 per cent and 75 per cent quartiles as boundary values between the three ranges. As in the case of pH, the influence of Sector IV was felt less, and Sector I more, as pollutant concentrations decreased.

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^{\pm} and $NO_3^{\pm}-N$ concentrations measured in precipitation with air trajectories from each major source sector (Table 7). For the most part, the average pH decreased and the average excess sulphate and nitrate concentrations increased as the sector emission levels increased (see Table 1).

		рH	[XSO4 ⁼] mg·l ⁻¹	[NO ₃ ⁻ -N] mg·l ⁻¹	
Sector	I	4.91	0.48	0.062	
Sector	II	4.82	0.45	0.084	
Sector	III	4.52	0.81	0.266	
Sector	IV	4.32	1.60	0.341	

Table 7: Precipitation-Weighted Average pH, and Average XSO4[‡] and NO3⁺-N Concentrations in Precipitation by Major Source Sector at Kejimkujik, N.S., 1992

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). Α study of large deposition episodes occurring at Kejimkujik during the early part of the decade (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 during the year. It is likely that this episodic events characteristic occurs every year because the 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 1992 again showed this episodic characteristic. After ranking the 187 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 about 5% of the precipitation days. Tables 8 to 10 show the data for the top 5 episodes for each chemical. Nearly all of these top episodes were common to all three chemicals.

For H^{*} deposition, about 5% (10 out of 185) of the days with pH measurements were responsible for over 30% of the measured H^{*} deposition for the year. For XSO_4^{\pm} and NO_3^{-} , between 4% and 5% (8 out of 187 and 9 out of 187, respectively), were responsible for over 30% of the measured wet deposition for each chemical. Therefore, the deposition at Kejimkujik in 1992 was not only episodic but, in fact, qualified as highly episodic for XSO_4^{\pm} and NO_3^{-} . Highly episodic deposition (when less than 5% of the annual precipitation days are episode days) usually occurs in areas remote from the largest source regions (Pasquill and Smith, 1983).

A seasonal analysis of the large deposition episodes showed a spring bias in 1992, especially for the hydrogen and sulphate ions. Between 33% and 50% of the large deposition events occurred in the spring months (April to June) depending on the ion (33% for nitrate, 50% for hydrogen ion and 50% for sulphate). Moreover, the month of June accounted for 75% of large deposition spring events. A spring - summer bias was noted for 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 at Kejimkujik (Beattie and O'Brien, 1992b), as well as at Harcourt, N.B. (Beattie *et al.*, 1993).

There were also biases in precipitation type, with 9 out of 10 (90%) of the large deposition episodes for hydrogen ion being rain events, while the remaining event consisted of both rain and snow (a "mixed" event). Seven out of 8 (88%) of the large deposition events for excess sulphate were rain events, with one mixed event. For nitrate, 7 of the 9 episodes (78%) 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 and 1991 (Beattie and O'Brien, 1992a and 1992b; Beattie et al., 1993).

Table 8:	The Five Largest Wet H ⁺ Deposition Events
	at Kejimkujik, N.S. During 1992

		Preci	oitation			Wet H+ I	Deposition
Event #	Date M/D	Type (1)	Amount (mm)	Upwind Sectors (2)	рН	Amount mg·m ⁻²	% of Total (3)
1	10/29	R	9.2	IV	3.80	1.46	4.7%
2	05/02	R	6.2	IV,III	3.63	1.45	4.7%
3	06/08	R	12.4	IV	3.97	1.33	4.3%
4	06/15	R	11.2	IV,I	3.94	1.29	4.1%
5	06/30	R	4.0	ĬV	3.66	0.88	2.8%
				Total:		6.40 (4)	20.6%

NOTES:

(1) Precipitation Type: R = Rain

(2) Upwind sectors are defined in Table 1.

(3) Total measured deposition was 31.1 mg m⁻².

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

Table 9: The Five Largest Wet XSO4⁼ Deposition Events at Kejimkujik, N.S. During 1992

Event #	Date M/D	Preci Type (1)	<u>pitation</u> Amount (mm)	Upwind Sectors (2)	[XSO4 ⁼] mg·1 ⁻¹	Wet XSO Amount mg·m ⁻²	4 Deposition % of Total (3)
1	06/15	R	11.2	IV.I	5.36	60.0	5.6%
2	06/08	R	12.4	IV	4.52	56.0	5.3%
3	05/02	R	6.2	IV,III	7.80	48.4	4.5%
4	06/30	R	4.0	iv	11.62	46.5	4.4%
5	10/29	R	9.2	IV	4.30	39.6	3.7%
<u> </u>			<u> </u>	Total		250.5	23.6% (4)

NOTES:

(1) Precipitation Type: R = Rain

(2) Upwind sectors are defined in Table 1.

(3) Total measured deposition was 1062.9 mg·m⁻².

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

		Preci	pitation			Wet NO3	Deposition
Even	t Date	Туре	Amount	Upwind	[NO3 -N]	Amount	% of Total
#	M/D	(1)	(mm)	Sectors	mg·1 ⁻¹	mg·m ⁻²	(3)
	·			(2)	-	-	
1	10/29	R	9.2	IV	1.44	58.7	6.3%
Ż	05/02	R	6.2	IV,III	1.70	46.7	5.0%
3	06/15	R	11.2	IV,I	0.84	41.7	4.5%
4	06/08	R	12.4	IV	0.55	30.2	3.3%
5	02/11	S	5.6	IV	1.09	27.0	2.9%
<u> </u>		<u></u>	T	otal :		204.3	22.0%
NOTE	s:						
(1)	Precipita	ation	Type: R	t = Rain			
(2) (3)	Upwind se Total mea	ectors asured	are def deposit	ined in the second s	Table 1. 928.9 mg	m ⁻² .	

Table 10: The Five Largest Wet NO3 Deposition Events at Kejimkujik, N.S. During 1992

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 (Figure 20) 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 11) and compared to the pollutant Ten out of 187 precipitation days (about 5.3%) episodicity. accounted for approximately 30% of the total precipitation amount of 1153.9 mm. Thus, though precipitation events were episodic in nature, they were not quite as episodic as excess sulphate and nitrate depositions. 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 by the fact that only one of the precipitation events which comprised the top 30% of precipitation amount was also a pollutant episode.

Air Trajectory to Kejimkujik, N.S. for the Largest Combined $(H^{+}, XSO_{4}^{-}, and NO_{3})$ Deposition Episode During 1992 (June 15)



Table	11:	The	Five	Larges	st Pre	ecipitat	ion	Events
		at 1	Kejimk	ujik,	N.S.	During	1992	2

Ē	vent #	Date M/D	Type (1)	Amount (mm)	% of Tot (2)	al	
	1	03/25	 R	76.4	6,6%		
	2	01/23	M	43.8	3.8%		
	3	11/13	R	38.6	3.3%		
	4	01/05	R	33.0	2.9%		
	5	12/17	R	32.8	2.8%		
	Tota	1:		224.6	19.5%	(3)	

NOTES:

 Precipitation Type: R = Rain M = Mixed Rain and Snow
Total measured precipitation was 1153.9 mm.
Small difference between sum and Total is due to roundoff.

3.6 Trends over Period of Record

Figure 21 shows the average annual pH of precipitation received at Kejimkujik from 1980 to 1992. The least acidic of the thirteen 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 is shown in Table 12.

Table 12: Average Precipitation-Weighted pH and Range of pHs in Precipitation at Kejimkujik, N.S., During 1981-1992

PH	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992
Min	3.60	3.40	3.60	3.15	3.49	3.31	3.55	3.41	3.36	3.40	3.52	3.04
Mean	4.60	4.56	4.71	4.66	4.40	4.44	4.58	4.59	4.44	4.63	4.58	4.57
Max	7.40	7.30	6.00	6.50	5.48	5.39	5.57	5.63	5.50	5.82	5.50	5.47

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). Figure 22 and Table 13 illustrate that the largest annual wet hydrogen ion deposition from





1980 to 1992 (0.53 kg·ha⁻¹) occurred in 1986. The deposition in recent years has not fallen to the low levels observed in 1983 (0.26 kg·ha⁻¹) and 1984 (0.29 kg·ha⁻¹).

Annual wet deposition amounts for excess sulphate and nitrate are shown in Figure 23 and Table 13. For excess sulphate, the annual wet deposition from 1980-1992 varied from a high in 1981 of 20.2 kg·ha⁻¹, to a low in 1983 of 10.6 kg·ha⁻¹ (with the 1992 deposition of 10.8 kg·ha⁻¹ very close to the low mark). For nitrate, the annual wet deposition varied from a high in 1989 of 14.4 kg·ha⁻¹ to a low in 1983 of 5.4 kg·ha⁻¹. (For Figures 21 to 23, and Tables 12 and 13, 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 1988-1991 data from A.E.S. (1989; 1990; 1991; 1992).)

Table 13: Annual H⁺, XSO₄[±] and NO₃⁻ Depositions, and Annual Precipitation Kejimkujik, N.S., 1980-1992

Year	Dep H⁺	osition (XSO4 ⁼	kg·ha ⁻¹) NO3 ⁻		Precipitation (mm)					
1980	0.43	18.0	10.0		1190.6					
1981	0.41	20.2	10.1		1625.2					
1982	0.35	14.6	7.8		1260.2					
1983	0.26	<u>10.6</u>	<u>5.4</u>		1344.1					
1984	0.29	14.7	8.2		1315.0					
1985	0.51	17.0	12.1		1298.7					
1986	0.53	17.0	11.6		1454.0					
1987	0.33	12.3	9.3		1238.2					
1988	0.37	14.0	10.1		1428.2					
1989	0.47	16.7	14.4		1304.8					
1990	0.38	13.8	10.7		1607.3					
1991	0.38	14.8	10.8		1436.8					
1992	0.32	10.8	9.4		1177.0					
Average	0.39	15.0	10.0		1360.0					
NOTE: underlir	Maxima ned.	in each	column ar	e typed i	n bold; mini	ma are				

Although similar cycles occur in the annual deposition amounts for the three ions at Kejimkujik, the year with the largest wet deposition is different for each (1986 for H⁺, 1981 for $XSO_4^{=}$, 1989 for NO_3^{-}). The lowest annual wet deposition totals occurred in 1983 for all three ions. 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 1992 to the average depositions for the last five years of the period showed that the average annual excess sulphate deposition decreased by over 10% from the first five years to the last five years of the period, whereas the average annual nitrate deposition increased by over 33%. An examination of yearly precipitation totals for the period shows that there was little change (a 3% increase) from the first five years to the last five years.

Statistical 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, Excess sulphate concentrations decreased monotonically, 1994). though not linearly, during the period, with a decrease of over 40% However, the average precipitation-weighted concentration noted. for the five years 1980 to 1984 was 1.16 mg⁻¹⁻¹, whereas the average concentration for the five year period 1988 to 1992 was 1.01 mg·l⁻¹, representing a reduction of only about 13%. Nitrate concentrations decreased during the early 1980's, then increased to reach a maximum during the late 1980's, exhibiting a trend similar to hydrogen ion concentration. Average precipitation-weighted nitrate concentration for the five years 1980 to 1984 was 0.14 mg·l⁻¹, whereas the average concentration for the five year period 1988 to 1992 was 0.18 mg⁻¹⁻¹, representing an increase of about 29%.

Visual evidence for a decrease in excess sulphate concentration at Kejimkujik is shown in Figure 24, where the annual average precipitation-weighted excess sulphate concentration is plotted for the 13-year period 1980-1992, 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 25) shows evidence for a decrease in the early 1980's and an increase since then.

The relative importance of excess sulphate versus nitrate deposition at Kejimkujik (Figure 26) 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 1988 to 1992 (1.27). The largest annual ratio (2.00) occurred in 1981 and the smallest (1.15) occurred in 1992. Based on ion equivalents, excess sulphate contributed 2.5 times as much as nitrate to precipitation acidity in 1981, but only 1.5 times as much in 1992. This change at Kejimkujik may be the result of a decrease in the ratio of Eastern North American SO₂ to NO_x emissions during the period due to a much larger reduction in anthropogenic SO₂ emissions than in NO_x emissions (Sirois and Summers, 1989; RMCC, 1990).

Figure 24 Annual Excess Sulphate Concentration in Precipitation at Kejimkujik, N.S. 1980-1992





Figure 25 Annual NO3-N Concentration in Precipitation at Kejimkujik, N.S. 1980-1992



4. Conclusions

The average annual precipitation-weighted pH for 1992 at Kejimkujik National Park in southwestern Nova Scotia was 4.57, with the most acidic seasonal average in spring and the least acidic in the winter. The most acidic event of the year (pH of 3.04) was 269 times more acidic than the least acidic event (pH of 5.47). About 17% of the events were highly acidic (pH \leq 4.0), contributing 39% of the total annual hydrogen ion deposition. In comparison with the previous year, Kejimkujik experienced a 16% decrease in H⁺ deposition primarily due to a 18% decrease in precipitation.

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 1992 at Kejimkujik was 10.8 kg ha⁻¹ and 9.4 kg ha⁻¹, respectively. Average annual precipitation-weighted concentrations were 0.92 mg l⁻¹ for excess sulphate and 0.18 mg l⁻¹ 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. 4% or 5% depending on the chemical). The largest daily deposition occurred on June 15, when 0.6 kg ha⁻¹ of excess sulphate (almost 6% of the annual total) and 0.09 kg ha⁻¹ of nitrate (almost 5% of the annual total) were deposited. The pollutant source region for this large deposition episode was the sector with the largest emissions (eastern U.S.A. and southern Ontario).

The least acidic year from 1980 to 1992 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⁻¹ to a low in 1983 of 10.6 kg ha⁻¹. For nitrate, the annual wet deposition varied from a high in 1989 of 14.4 kg ha⁻¹ to a low in 1983 of 5.4 kg ha⁻¹. The large variability in the annual excess sulphate deposition data all but obscured the significant decreasing trend (more than a 40% decrease) found 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 decrease in the ratio of Eastern North American SO₂ to NO, emissions during the decade.

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APPENDIX A Example of Acid Rain Report



ACID RAIN

The reference map (left) shows the locations of sampling sites, where the acidity of precipitation is monitored. All are operated by Environment Canada except Dorset (*), which is a research station operated by the Ontario Ministry of the Environment. The map also shows the approximate areas (shaded), where SO₂ and NO_x emissions are greatest.

The table below gives the weekly report summarizing the acidity (or pH) of the acid rain or snow that fell at the collection sites, and a description of the path travelled by the moisture laden air. Environmental damage to lakes and streams is usually observed in sensitive areas regularly receiving precipitation with pH readings less than 4.7, while pH readings less than 4.0 are serious.



SITE	day	pH a	imo	unt	AIR	PATH TO SITE
						June 14 to 20, 1992
Longwoods	17	4.3	24	R	• • • •	. Western Ohio, Indiana, Kentucky
Dorset *	17	3.9	3	R		. Southern Ontano, western Ohio, Indiana
	18	4.7	15	R		. Southern Ontano, northern Ohio, northern Indiana
	20	4.6	6	R	• • • •	. Northwestern Quebec
Chalk River					••••	. Data not available this week
Sutton	20	4.7	10	R	• • • •	. Northern New York
Montmorency	19	4.2	2	R		. Southern Quebec, northern New York
·	20	4.8	22	R		. Southern Quebec, Vermont, New Hampshire
Kejimkujik	15	3.9	11	R		. Southern New England
	20	4.9	12	R		Atlantic Ocean

R= rain (mm), S = snow (cm), M = mixed rain and snow (mm)

APPENDIX B

Precipitation Chemistry Data Set

Kejimkujik, N.S.

1992

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DATE	PCP	PCP	нa	SECTOR	H+ DEPN	XSO4	XSO4	NO3-N	NO3-N
	TYP	AMT	1		(mg/m**2)	CONC	DEPN	CONC	DEPN
		(mm)			(),	(mg/l)	(mg/m**2)	(mg/l)	(mg/m**2)
		<u> </u>							
01/04/92	R	0.8	4.43		0.0298	1.80	1.44	0.10	0.08
01/05/92	R	33.0	5.36	1	0.1452	0.08	2.64	0.02	0.66
01/06/92	R	0.8	5.20		0.0050	0.24	0.19	0.04	0.03
01/07/92	М	0.4	3.97		0.0429	5.75	2.30	0.42	0.17
01/08/92	S	0.8	4.75		0.0142	1.04	0.83	0.08	0.06
01/09/92	Μ	5.4	4.41	4 1	0.2101	1.08	5.83	0.32	1.73
01/10/92	Μ	0.4	4.42		0.0152	1.37	0.55	0.30	0.12
01/11/92	S	3.0	4.64	32	0.0687	0.65	1.95	0.22	0.66
01/13/92	Μ	0.6	3.37		0.2560	10.15	6.09	5.10	3.06
01/14/92	R	25.2	5.07	1	0.2142	0.28	7.06	0.04	1.01
01/15/92	S	3.0	4.58	43	0.0789	1.50	4.50	0.23	0.69
01/16/92	S	2.2	4.73	3	0.0409	0.34	0.75	0.23	0.51
01/17/92	S	1.4	4.08	43	0.1165	0.71	0.99	1.10	1.54
01/18/92	S	3.0	4.68	3	0.0627	1.15	3.45	0.47	1.41
01/19/92	S	2.0	4.54	3	0.0576	0.76	1.52	0.30	0.60
01/20/92	S	1.2	4.35	43	0.0536	0.23	0.28	0.55	0.66
01/21/92	S	1.4	4.17	3	0.0946	0.54	0.76	0.81	1.13
01/22/92	S	1.6	4.88	3	0.0211	0.25	0.40	0.13	0.21
01/23/92	Μ	43.8	4.86	1	0.6044	0.56	24.53	0.05	2.19
01/24/92	S	1.4	4.25	43	0.0787	2.49	3.49	0.86	1.20
01/25/92	S	0.6	4.77		0.0102	0.48	0.29	0.27	0.16
01/26/92	S	0.4	4.30		0.0200	0.57	0.23	0.52	0.21
01/30/92	R	0.8	4.27		0.0430	1.05	0.84	0.62	0.50
01/31/92	Μ	6.4	4.45	21	0.2272	1.54	9.86	0.16	1.02
02/01/92	S	24.4	5.21	2	0.1513	0.05	1.22	0.03	0.73
02/02/92	S	1.8	4.63	21	0.0421	0.58	1.04	0.18	0.32
02/03/92	S	0.4	4.50		0.0126	1.46	0.58	0.06	0.02
02/04/92	S	3.2	4.44	2	0.1162	0.27	0.86	0.42	1.34
02/05/92	S	16.0	5. 19	2	0.1040	0.16	2.56	0.03	0.48
02/07/92	S	2.6	5.06	1	0.0226	0.14	0.36	0.08	0.21
02/08/92	S	10.6	5.47	21	0.0360	0.00	0.00	0.03	0.32
02/09/92	S	3.2	4.34	43	0.1462	0.67	2.14	0.52	1.66
02/11/92	S	5.6	4.04	4	0.5107	1.21	6.78	1.09	6.10
02/12/92	S	0.4	4.17		0.0270	0.98	0.39	0.70	0.28
02/13/92	S	4.4	4.60	4	0.1104	0.87	3.83	0.11	0.48
02/14/92	S	0.2	4.42		0.0076	1.74	0.35	0.17	0.03
02/15/92	Μ	32.2	5.03	1	0.2995	0.27	8.69	0.02	0.64
02/16/92	Μ	4.2	4.83	1	0.0622	0.72	3.02	0.03	0.13
02/18/92	R	6.2	5.00	1	0.0620	0.45	2.79	0.05	0.31

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DATE	PCP	PCP	pН	SECTOR	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)
								, ,	
02/19/92	R	1.0	5.31	1	0.0049	0.14	0.14	0.01	0.01
02/21/92	S	6.4	4.52	43	0.1933	0.23	1.47	0.32	2.05
02/22/92	S	2.6	3.80	4	0.4121	4.13	10.74	1.62	4.21
02/23/92	S	0.4	4.69		0.0082	1.54	0.62	0.41	0.16
02/25/92	Μ	10.8	4.97	1	0.1156	0.36	3. 89	0.05	0.54
02/26/92	S	4.3	5.23	1	0.0254	0.09	0.39	0.03	0.13
02/27/92	S	1.2	4.33	4	0.0562	1.65	1.98	0.42	0.50
02/28/92	R	4.8	3.91	4 1	0.5904	3.99	19.15	1.27	6.10
02/29/92	S	14.8	4.59	32	0.3804	0.56	8.29	0.23	3.40
03/01/92	S	1.2	4.40	43	0.0478	1.21	1.45	0.35	0.42
03/03/92	S	0.8	4.26		0.0440	0.64	0.51	0.65	0.52
03/07/92	R	17.2	4.78	1	0.2855	0.68	11.70	0.05	0.86
03/08/92	R	5.4	4.77	1	0.0918	0.77	4.16	0.06	0.32
03/10/92	R	0.2	-9.00		-9.0000	0.84	0.17	-0.05	0.01
03/11/92	R	14.2	4.86	43	0.1960	0.56	7.95	0.08	1.14
03/12/92	S	2.0	3.90	4	0.2518	2.11	4.22	1.83	3.66
03/13/92	S	1.2	4.20	4	0.0757	1.53	1.84	0.78	0.94
03/14/92	S	1.0	4.46	43	0.0347	0.49	0.49	0.41	0.41
03/17/92	S	0.8	3.97		0.0858	2.38	1.90	1.71	1.37
03/22/92	S	0.6	4.80		0.0095	0.33	0.20	0.20	0.12
03/23/92	S	2.0	5.17	4 1	0.0136	0.26	0.52	0.05	0.10
03/25/92	R	76.4	5.24	1	0.4431	0.32	24.45	0.03	2.29
03/28/92	R	6.0	5.16	1	0.0414	0.52	3.12	0.10	0.60
03/29/92	R	11.2	4.62	1	0.2688	1.06	11.87	0.13	1.46
04/02/92	S	3.0	4.80	1	0.0474	0.87	2.61	0.11	0.33
04/03/92	S	8.0	4.80	2	0.1264	0.57	4.56	0.09	0.72
04/05/92	R	0.2	4.94		0.0023	0.72	0.14	-0.05	0.01
04/08/92	R	0.2	3.92		0.0240	3.36	0.67	2.06	0.41
04/09/92	R	1.3	3.97	3	0.1394	2.11	2.74	1.27	1.65
04/10/92	R	2.2	3.94	41	0.2526	3.85	8.47	0.78	1.72
04/11/92	S	19.6	4.57	32	0.5272	0.88	17.25	0.11	2.16
04/12/92	S	0.4	4.61		0.0098	1.50	0.60	0.25	0.10
04/14/92	S	0.2	4.27		0.0107	1.41	0.28	0.60	0.12
04/17/92	Μ	0.6	4.61		0.0147	1.18	0.71	0.15	0.09
04/22/92	R	3.7	4.42	1	0.1406	1.94	7.18	0.25	0.93
04/23/92	R	1.2	4.42	4	0.0456	1.55	1.86	0.20	0.24
04/24/92	М	1.4	4.40	32	0.0557	1.72	2.41	0.15	0.21
04/25/92	S	22.0	4.83	1	0.3256	0.49	10.78	0.08	1.76
04/26/92	S	7.0	4.38	21	0.2919	1.78	12.46	0.15	1.05

DATE	PCP	PCP	pН	SECTOR	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)
05/02/92	R	6.2	3.63	43	1.4533	7.80	48.36	1.70	10.54
05/05/92	S	4.4	4.89	1	0.0568	0.66	2.90	0.07	0.31
05/08/92	R	4.4	4.85	1	0.0620	0.72	3.17	0.13	0.57
05/09/92	R	12.6	4.75	1	0.2243	0.92	11.59	0.09	1.13
05/12/92	R	0.4	4.52		0.0121	1.59	0.64	0.11	0.04
05/13/92	R	2.0	4.70	1	0.0400	0.80	1.60	0.10	0.20
05/14/92	R	0.6	4.66		0.0131	0.79	0.47	0.18	0.11
05/18/92	R	10.2	4.39	4	0.4151	1.62	16.52	0.17	1.73
05/25/92	R	0.2	3.04		0.1824	38.28	7.66	5.69	1.14
05/26/92	R	0.2	3.61		0.0491	9.10	1.82	1.80	0.36
05/27/92	R	0.4	3.82		0.0606	6.32	2.53	0.62	0.25
06/01/92	R	1.6	4.67	1	0.0342	0.59	0.94	0.13	0.21
06/02/92	R	4.8	4.94	1	0.0552	0.29	1.39	0.07	0.34
06/04/92	R	0.8	3.73		0.1490	11.19	8.95	1.23	0.98
06/06/92	R	12.6	4.73	1	0.2344	0.82	10.33	0.08	1.01
06/08/92	R	12.4	3.97	4	1.3293	4.52	56.05	0.55	6.82
06/14/92	R	0.4	3.65		0.0896	6.48	2.59	2.46	0.98
06/15/92	R	11.2	3.94	4 1	1.2858	5.36	60.03	0.84	9.41
06/20/92	R	12.4	4.83	1	0.1835	0.49	6.08	0.08	0.99
06/21/92	R	6.6	4.77	1	0.1122	0.52	3.43	0.11	0.73
06/22/92	R	5.8	4.62	1	0.1392	0.62	3.60	0.16	0.93
06/24/92	R	6.6	3.99		0.6752	3.42	22.57	0.63	4.16
06/25/92	R	3.8	4.87		0.0513	0.33	1.25	0.09	0.34
06/30/92	R	4.0	3.66	4	0.8752	11.62	46.48	0.92	3.68
07/04/92	R	6.6	4.92	1	0.0792	0.31	2.05	0.06	0.40
07/05/92	R	2.0	4.18	4	0.1322	1.88	3.76	0.79	1.58
07/06/92	R	2.8	3.91	4 1	0.3444	3.54	9.91	1.13	3.16
07/09/92	R	10.6	4.40	4 1	0.4219	0.95	10.07	0.28	2.97
07/12/92	R	3.0	4.17	4	0.2028	1.90	5.70	0.34	1.02
07/13/92	R	10.6	4.30	4	0.5311	2.71	28.73	0.20	2.12
07/14/92	R	3.8	5.07	4	0.0323	0.08	0.30	0.07	0.27
07/18/92	R	8.9	4.21	4 1	0.5491	2.38	21.18	0.47	4.18
07/19/92	R	0.8	3.81		0.1239	5.33	4.26	1.10	0.88
07/27/92	R	17.8	4.86	1	0.2456	0.61	10.86	0.07	1.25
07/28/92	R	2.4	4.85	4	0.0338	0.23	0.55	0.10	0.24
07/29/92	R	0.8	4.23		0.0471	⁻ 3.26	2.61	0.58	0.46
07/31/92	R	3.1	3.77	1	0.5264	7.84	24.30	0.94	2.91
08/01/92	R	10.0	4.57	1	0.2690	1.06	10.60	0.14	1.40
08/04/92	R	5.6	4.69	1	0.1142	0.81	4.54	0.14	0.78

DATE	PCP TYP	PCP AMT (mm)	pН	SECTOR	H+ DEPN (mg/m**2)	XSO4 CONC (mg/l)	XSO4 DEPN (mg/m**2)	NO3-N CONC (mg/l)	NO3-N DEPN (mg/m**2)
08/09/92	R	02	4.37		0.0085	0.23	0.05	0.07	0.01
08/10/92	R	4.6	4.01	1	0 1523	1.34	6.16	0.14	0.64
08/11/92	R	0.2	3.93	•	0.0235	4.37	0.87	0.91	0.18
08/15/92	R	0.2	4 23		0.0118	1.33	0.27	0.29	0.06
08/16/92	8	4.6	4 80	4	0.0727	0.48	2.21	0.11	0.51
08/17/92	R	14.8	5.42	4 1	0.0562	0.08	1.18	0.03	0.44
08/18/92	R	14.2	5 19	1	0.0923	0.15	2.13	0.02	0.28
08/27/92	R	0.4	4.61	•	0.0098	1.28	0.51	0.13	0.05
08/29/92	R	1.0	4.51		0.0309	1.33	1.33	0.26	0.26
09/01/92	R	0.2	4.84		0.0029	1.49	0.30	0.30	0.06
09/03/92	R	4.6	5.25	1	0.0258	0.23	1.06	0.06	0.28
09/04/92	R	15.8	5.20	1	0.0995	0.13	2.05	0.03	0.47
09/08/92	R	0.2	3.95		0.0224	5.51	1.10	0.58	0.12
09/09/92	R	0.2	3.20		0.1262	16.93	3.39	4.76	0.95
09/11/92	R	5.7	4.43	4 1	0.2120	1.04	5.93	0.24	1.37
09/19/92	R	7.2	3.97	4	0.7718	3.53	25.42	0.59	4.25
09/22/92	R	20.0	5.04	4 1	0.1820	0.33	6.60	0.03	0.60
09/23/92	R	1.2	4.65	4	0.0269	0.75	0.90	0.12	0.14
09/27/92	R	1.0	4.34	4	0.0457	1.50	1.50	0.31	0.31
09/29/92	R	5.8	4.53	4	0.1711	0.74	4.29	0.21	1.22
10/01/92	М	0.2	5.03		0.0019	0.68	0.14	-0.05	0.01
10/02/92	R	2.0	4.38	43	0.0834	2.06	4.12	0.60	1.20
10/10/92	R	21.6	5.21	1	0.1339	0.23	4.97	0.02	0.43
10/11/92	R	20.2	5.04	1	0.1838	0.29	5.86	0.05	1.01
10/14/92	R	1.6	4.13	3	0.1186	1.09	1.74	1.04	1.66
10/19/92	Μ	4.0	5.24	32	0.0232	0.14	0.56	0.02	0.08
10/21/92	R	23.2	5.15	1	0.1647	0.27	6.26	0.02	0.46
10/22/92	R	0.2	4.44		0.0073	1.62	0.32	0.16	0.03
10/25/92	R	6.2	4.58	1	0.1631	1.00	6.20	0.14	0.87
10/26/92	R	1.2	4.86	3	0.0166	0.49	0.59	0.09	0.11
10/29/92	R	9.2	3.80	4	1.4582	4.30	39.56	1.44	13.25
10/30/92	R	3.4	3.98	4	0.3560	4.16	14.14	0.51	1.73
11/01/92	Μ	1.0	4.78	2	0.0166	0.63	0.63	0.05	0.05
11/03/92	R	2.4	5.03	43	0.0223	0.26	0.62	0.04	0.10
11/04/92	R	4.8	4.20	1	0.3029	2.04	9.79	0.40	1.92
11/05/92	R	3.6	4.42	3	0.1368	1.49	5.36	0.16	0.58
11/06/92	R	0.8	4.54		0.0230	0.84	0.67	0.15	0.12
11/11/92	R	8.6	4.31	4	0.4214	1.88	16.17	0.20	1.72
11/12/92	R	5.0	4.77	41	0.0850	0.62	3.10	0.12	0.60

	DATE	PCP	PCP	pН	SEC	TOR	H+ DEPN	XSO4	XSO4	NO3-N	NO3-N
		TYP	AMT				(mg/m**2)	CONC	DEPN	CONC	DEPN
			(mm)					(mg/l)	<u>(mg/m**2)</u>	<u>(mg/l)</u>	(mg/m**2)
	11/13/92	R	38.6	5.42	1		0.1467	0.02	0.77	-0.01	0.19
	11/15/92	Μ	3.0	4.21	4		0.1851	1.76	5.28	0.54	1.62
	11/16/92	S	0.2	4.49			0.0065	0.40	0.08	0.38	0.08
	11/21/92	R	7.0	4.71	4		0.1365	0.71	4.97	0.11	0.77
	11/22/92	R	8.2	4.45	4		0.2911	0.92	7.54	0.23	1.89
	11/23/92	R	13.1	4.90	4	1	0.1651	0.50	6.55	0.02	0.26
	11/24/92	S	0.2	4.15			0.0142	7.78	1.56	0.55	0.11
	11/25/92	Μ	0.4	3 <i>.</i> 97			0.0429	3.40	1.36	0.43	0.17
	11/26/92	R	9.8	4.86	4	1	0.1352	0.55	5.39	0.06	0.59
	12/03/92	S	13.2	4.67	2	1	0.2825	0.63	8.32	0.13	1.72
	12/04/92	S	1.6	4.56	4	3	0.0440	0.29	0.46	0.35	0.56
·	12/05/92	S	11.4	4.90	4		0.1436	0.27	3.08	0.10	1.14
	12/06/92	S	3.8	4.46	З	2	0.1319	0.62	2.36	0.40	1.52
	12/07/92	S	5.2	4.34	4	3	0.2376	0.73	3.80	0.50	2.60
	12/08/92	S	0.8	4.35			0.0358	0.68	0.54	0.30	0.24
	12/09/92	S	0.2	4.71			0.0039	0.07	0.01	0.15	0.03
	12/11/92	S	0.2	-9.00			-9.0000	3.07	0.61	0.49	0.10
	12/16/92	R	0.4	3.78			0.0664	5.26	2.10	1.35	0.54
	12/17/92	R	32.8	4.92	4		0.3936	0.25	8.20	0.04	1.31
	12/18/92	R	0.2	4.09			0.0163	1.91	0.38	0.80	0.16
	12/19/92	R	4.4	4.60	4	1	0.1104	0.44	1.94	0.15	0.66
	12/20/92	Μ	17.0	4.72	4		0.3247	0.55	9.35	0.07	1.19
	12/22/92	S	0.4	4.07			0.0340	2.53	1.01	0.65	0.26
	12/23/92	R	0.2	3.34			0.0914	6.86	1.37	4.84	0.97
	12/24/92	S	4.8	4.32	З		0.2299	1.28	6.14	0.52	2.50
	12/25/92	S	5.0	4.58	4		0.1315	0.66	3.30	0.17	0.85
	12/26/92	S	2.4	4.69	3		0.0490	0.74	1.78	0.32	0.77
	12/28/92	R	0.2	4.17			0.0135	2.75	0.55	0.49	0.10
	12/29/92	М	17.4	4.91	1		0.2140	0.54	9.40	0.08	1.39
	12/30/92	S	1.4	4.21	4	1	0.0864	1.76	2.46	0.37	0.52
	12/31/92	Μ	3.2	4.46	4	1	0.1110	1.39	4.45	0.21	0.67

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

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Acid precipitation during 1992 at Kejimkujik, N.S. BEATTIE, BILLIE L

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