

ACID RAIN TENDENCIES DURING MAY, JUNE, AND JULY 1988 IN WEST LAFAYETTE, INDIANA

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ABSTRACT: Daily rainwater samples were collected from 29 April to 29 July 1988 at the Purdue University Agronomy Farm, six miles northwest of West Lafayette, Indiana. A total of 18 samples of 0.01 in. or more were collected; rainfall for the period was about 4.7 in., only 37% of normal. The average pH for the 18 samples was about 3.9, approximately 50 times more acidic than naturally-occurring rainfall. The average pH of 3.9 was lower (i.e., more acidic) than the 5-year average of about 4.2. Heavy rainfalls tended to be more acidic than light rainfalls, as the six heaviest rainfalls (pH = 3.9) were six times more acidic than the six lightest rainfalls (pH = 4.7). A strong correlation also existed between wind direction and acidity. Rainwater tended to be more acidic when the upper-air winds were from the south, southeast, or southwest prior to the rain event. These directions correspond to the regions of heavy coal burning by power plants in the Ohio Valley.

INTRODUCTION

Acid deposition has become an increasingly publicized issue in recent years. Extensive scientific and economic studies have been undertaken to examine the cause of acid deposition and the economic effects of proposed solutions. This study attempts to identify factors affecting the acidity of rainfall measured at one rural location in north-central Indiana during a three-month period in the spring and early summer of 1988.

In this study, only wet deposition, or "acid rain", was analyzed. As opposed to dry deposition, which involves deposition of sulfur and nitrogen oxide gases and particulates, wet deposition includes only those acid compounds deposited in liquid form (General Accounting Office, 1984). Sulfur dioxide (SO₂) and nitrogen oxides (NO and NO₂) are emitted by coal-fueled power plants, smelters, vehicles, and other sources. These emissions combine with water vapor in the air to create sulfurous and sulfuric acids (H₂SO₃ and H₂SO₄) as well as nitrous and nitric acids (HNO₂ and HNO₃). These acid compounds are then transported by upper-air winds and return to earth during rainfall. Naturally-occurring rainfall, i.e., rainfall without anthropogenic contaminants, is normally slightly acidic (hydrogen ion activity [H⁺] = 2.51 × 10⁻⁶ or, equivalently, pH = 5.6) due primarily to the creation of carbonic acid from the combination of carbon dioxide and water vapor (Liken, *et al.*, 1979). However, average annual pH values of 4.2 or less have become common this decade in the eastern half of the U.S. (Mohnen, 1988).

TABLE 1. Record of rainfalls (≥ 0.01 in.).

Date	Time (GMT)	Amount (in.)	[H+]	pH	Rate (in./hr.)
8 May	2030-2130	0.05	1.55×10^{-5}	4.81	0.05
9 May	0430-0630	0.24	6.61×10^{-5}	4.18	0.12
10 May	2115-2130	0.02	1.45×10^{-6}	5.84	0.04
13 May	2030-2230	0.20	1.15×10^{-4}	3.94	0.10
23 May	2000-2100	0.27	1.25×10^{-4}	3.90	0.07
	0400-0600				
	1300-1400				
24 May	2300-0000	0.20	6.46×10^{-5}	4.19	0.04
	0100-0300				
	0800-0900				
9 June	0100-0400	0.13	3.24×10^{-5}	4.49	0.04
29 June	1100-1400	0.05	7.24×10^{-6}	5.14	0.02
30 June	1600-2000	0.08	1.86×10^{-4}	3.73	0.02
11 July	1500-1600	1.10	3.80×10^{-4}	3.42	0.28
	1930-2130				
	2300-0000				
14 July	1430-1500	0.02	2.95×10^{-7}	6.53	0.04
17 July	2230-2245	0.01	1.15×10^{-6}	5.94	0.04
18 July	1200-1245	0.03	6.61×10^{-7}	6.18	0.04
19 July	1515-1615	1.98	3.31×10^{-5}	4.48	0.49
	2300-0200				
20 July	1100-1500	0.13	1.74×10^{-5}	4.76	0.03
21 July	1500-1930	0.04	6.76×10^{-5}	4.17	0.01
22 July	2030-2130	0.15	3.63×10^{-5}	4.44	0.15
25 July	1330-1500	0.03	6.03×10^{-5}	4.22	0.02

For this study, rainfall samples were collected from 29 April to 29 July 1988 at the Purdue University Agronomy Farm, located 6 miles northwest of West Lafayette, Indiana (at $40^{\circ} 28' N$, $87^{\circ} 00' W$; elevation = 705 ft.). The samples were collected using a Geochem Metrics Model 301 Automatic Sensing Wet/Dry Precipitation Collector, the same type of instrument which has been used by the National Atmospheric Deposition Program (NADP) at the Agronomy Farm for several years. This device positions a lid over the rain-collecting bucket when no rain is falling and so prevents alkaline material, such as calcareous dust, from entering the bucket and skewing results during long dry spells. Comparison of the catch in this collector with a nearby reference raingage (which also recorded the times each rain event began and ended) suggests that the collection efficiency of the Geochem Metrics Model 301 is no less than 80%.

Samples were collected at 15Z each day and represent the total accumulation from rain event(s) during the previous 24 hours. Each sample was taken immediately to a laboratory in the Department of Agronomy on the main campus of Purdue University to determine its pH using a Fisher Accumet pH Meter Model 630. The equipment for the analysis was the same as that used by NADP. Comparison of the daily samples for 1988 with the corresponding NADP weekly sample taken at a nearby collector (of the same type as used here) indicated that no large difference in acidity resulted from use of the two different sampling intervals.

Wind patterns were analyzed to determine whether relationships exist between wind direction and acidity of associated rainfalls. In addition, NADP data

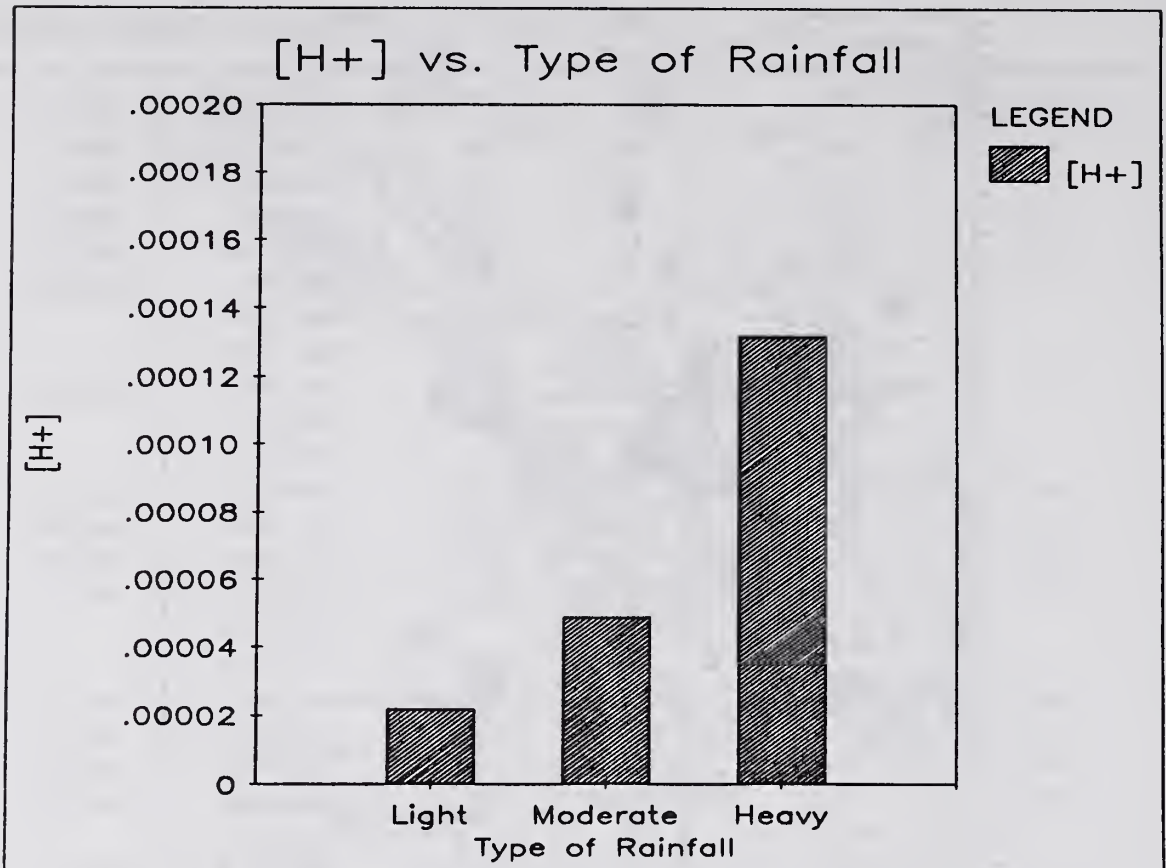


FIGURE 2. Variation in acidity according to amount of rainfall in an event.

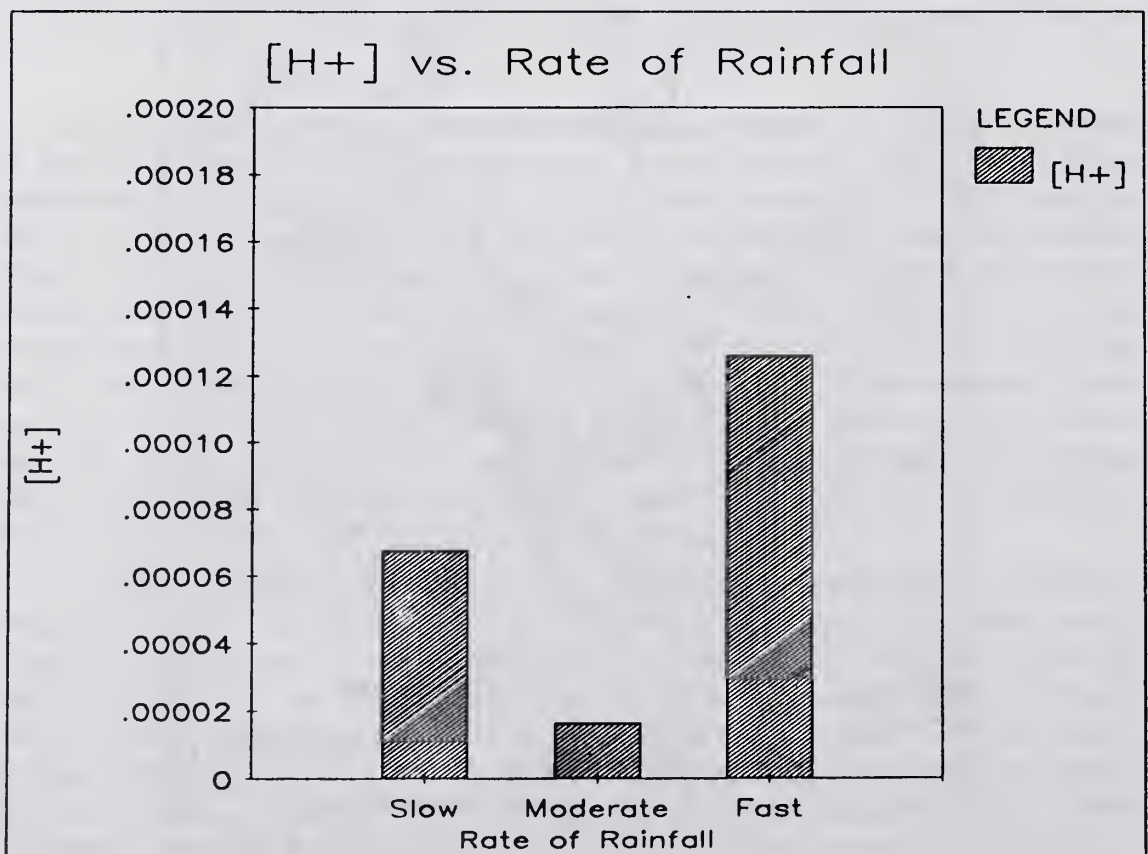


FIGURE 3. Variation in acidity according to rate of rainfall in an event.

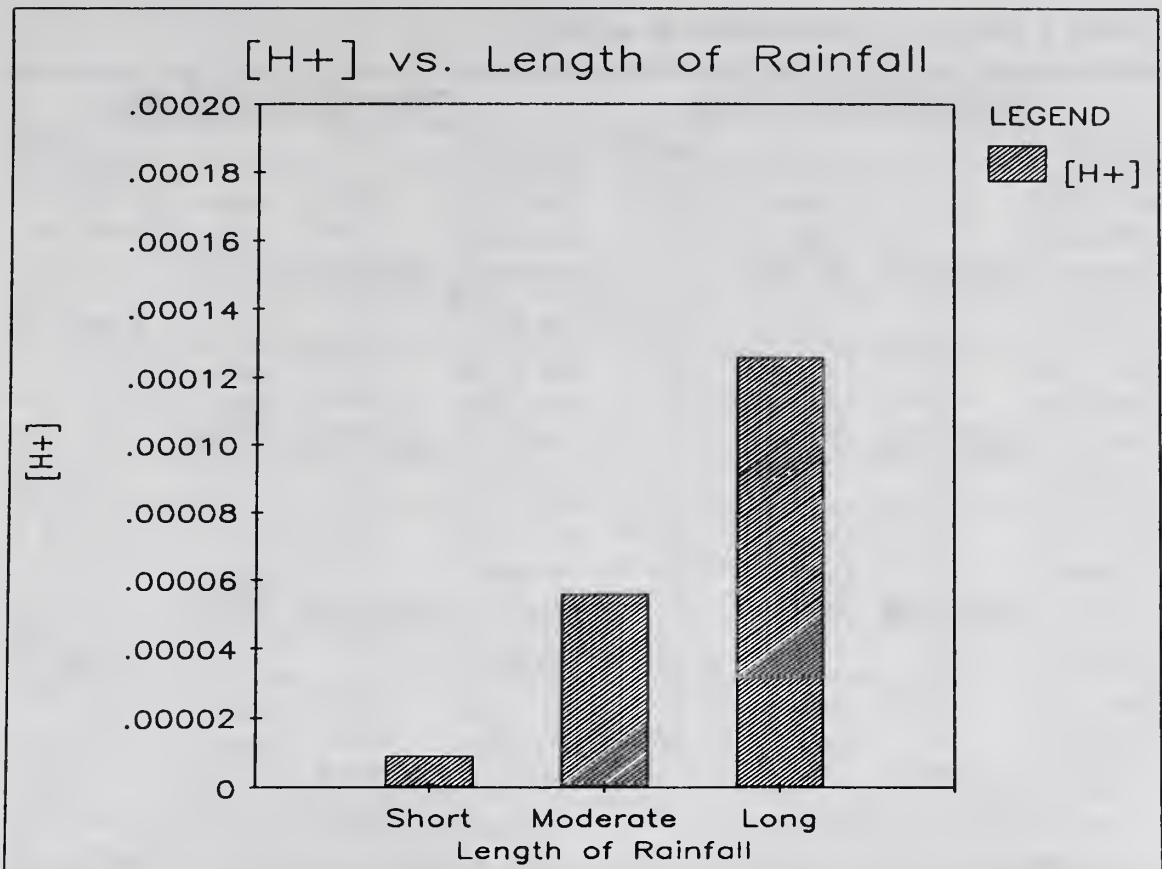


FIGURE 4. Variation in acidity according to length of rainfall in an event.

from the previous five years at the site were obtained and compared with data for this period in an effort to identify short term trends in rainfall acidity.

DATA AND ANALYSIS

Summary of rainwater samples. Samples from 18 rainfalls of 0.01 in. or more were obtained during the collection period. The date, time of production of each sample, rainfall amount, and pH are listed in Table 1. For use in later figures, hydrogen ion activity $[H^+]$ is also shown.

The average sample was 0.26 in. of rainwater with a pH of 3.9, about 50 times more acidic than naturally-occurring rainfall. (This is a weighted average and so is dominated by the two heavy rainfalls on 11 July and 19 July.) The most acidic rainfall occurred on 11 July (pH = 3.4), and the least acidic rainfall occurred on 14 July (pH = 6.5). Notice that the most acidic and least acidic samples for the entire collection period resulted from successive rain just 3 days apart, reflecting the high variability in acidity that occurred among events. The range of hydrogen ion activity from these two rainfalls represents about a 1300-fold difference in acidity.

Source regions. Figure 1 shows the distribution of sulfur dioxide and nitrogen oxide emissions in eastern North America. Recent studies indicate that in eastern North America sulfur dioxide is responsible for approximately two-thirds of the acidity in wet deposition, with nitrogen oxides contributing most of the remainder (General Accounting Office, 1984). Thus, sulfur dioxide appears to be the more important factor in wet deposition. Furthermore, a recent National

TABLE 2. Effect of wind direction on acidity.

24-48 Hours Before Rain			0-24 Hours Before Rain		
850 mb:	[H [±]]	Events	850 mb:	[H [±]]	Events
SW, S, SE:	9.33E-5	5	SW, S, SE:	1.12E-4	6
OTHERS :	4.79E-5	9	OTHERS :	4.47E-5	7
AMOUNT GREATER:	X 1.95		AMOUNT GREATER:	X 2.51	
700 mb:	[H [±]]	Events	700 mb:	[H [±]]	Events
SW, S, SE:	8.32E-5	6	SW, S, SE:	1.05E-4	7
OTHERS :	4.37E-5	9	OTHERS :	3.55E-5	7
AMOUNT GREATER:	X 1.90		AMOUNT GREATER:	X 2.96	
500 mb:	[H [±]]	Events	500 mb:	[H [±]]	Events
SW, S, SE:	1.20E-4	4	SW, S, SE:	1.41E-4	5
OTHERS :	3.72E-5	11	OTHERS :	3.09E-5	9
AMOUNT GREATER:	X 3.23		AMOUNT GREATER:	X 4.56	
300 mb:	[H [±]]	Events	300 mb:	[H [±]]	Events
SW, S, SE:	1.62E-4	3	SW, S, SE:	1.35E-4	5
OTHERS :	2.82E-5	10	OTHERS :	3.02E-5	8
AMOUNT GREATER:	X 5.74		AMOUNT GREATER:	X 4.47	
AVERAGE AMOUNT GREATER (ALL PRESSURE LEVELS): X 3.21			AVERAGE AMOUNT GREATER (ALL PRESSURE LEVELS): X 3.63		

Academy of Sciences report (1983) indicated that a linear relationship exists between the size of SO₂ emissions and the acidity of rainfall. Map A clearly shows that regions in southeastern Missouri, southwestern Illinois, and southern and eastern Indiana produce large amounts of SO₂ (Kurtz, *et al.*, 1984). These regions are located to the southwest, south, and southeast of the Purdue Agronomy Farm, respectively, and would be expected to contribute most heavily to wet deposition at this location, if wind strength and persistence were sufficient to transport acid compounds to the collection site. Within these southwest, south, and southeast sectors, the high SO₂-emission regions (black) are from 180 to 270 miles from the collection site. For acid compounds from these source regions to reach the collection site, winds need to average only 3-5 knots during the 48 hours prior to rainfall or 7-10 knots during the 24 hours prior to rainfall.

Horizontal transport. An analysis of wind directions and wind speeds occurring prior to each rainfall event was made at four levels in the atmosphere: 850 mb, 700 mb, 500 mb, and 300 mb. Wind data were not obtainable for one or more levels for each of the 18 rainfall events, so that the number of occurrences of wind analyzed at each level is less than the number of rainfall events.

The results presented in Table 2 show the effect of wind direction prior to a rain event on hydrogen ion activity in rainfall. Note particularly the differences in acidity associated with winds from the southwest (202.5°-247.5°), south (157.5°-202.5°), and southeast (112.5°-157.5°) as compared to winds from all other directions. The columns represent winds occurring between 24 to 48 hours before the rain event (48 hours before collection time) and winds up to 24 hours before the

TABLE 3. Effect of wind speed on acidity.

24-48 Hours Before Rain			0-24 Hours Before Rain		
850 mb:	[H [±]]	Events	850 mb:	[H [±]]	Events
< 10 KNOTS:	8.71E-5	8	< 10 KNOTS:	1.17E-4	6
> 10 KNOTS:	3.16E-5	6	> 10 KNOTS:	4.07E-5	7
AMOUNT GREATER:	X 2.75		AMOUNT GREATER:	X 2.87	
700 mb:	[H [±]]	Events	700 mb:	[H [±]]	Events
< 15 KNOTS:	7.94E-5	9	< 15 KNOTS:	5.89E-5	6
> 15 KNOTS:	2.88E-5	6	> 15 KNOTS:	7.94E-5	8
AMOUNT GREATER:	X 2.76		AMOUNT GREATER:	X .74	
500 mb:	[H [±]]	Events	500 mb:	[H [±]]	Events
< 20 KNOTS:	1.00E-4	7	< 20 KNOTS:	1.05E-4	6
> 20 KNOTS:	2.40E-5	8	> 20 KNOTS:	4.37E-5	8
AMOUNT GREATER:	X 4.17		AMOUNT GREATER:	X 2.41	
300 mb:	[H [±]]	Events	300 mb:	[H [±]]	Events
< 25 KNOTS:	1.07E-4	5	< 25 KNOTS:	1.51E-4	4
> 25 KNOTS:	2.82E-5	8	> 25 KNOTS:	3.47E-5	9
AMOUNT GREATER:	X 3.79		AMOUNT GREATER:	X 4.35	
AVERAGE AMOUNT GREATER (ALL PRESSURE LEVELS): X 3.37			AVERAGE AMOUNT GREATER (ALL PRESSURE LEVELS): X 2.59		

rain event (24 hours before the collection time). For every atmospheric level at both time intervals, the results show that the average acidity of rainfall was greater when winds were from the southwest, south, or southeast prior to the rainfall. The data show that on the average, when winds were from these directions 24 to 48 hours before rainfall, the hydrogen ion activity was about 3.2 times greater than when winds were from some other direction. For winds up to 24 hours before rainfall, a similar trend emerges, with acidity about 3.6 times greater with winds from the southwest, south, or southeast than that with winds from other directions.

In addition to wind direction, the acidity of rainfall was also correlated with wind speeds aloft, independent of the associated wind directions. Table 3 shows the effect of wind speed on acidity for the four pressure levels. For each pressure level, the wind speeds associated with the rain events were split into two nearly equal-size categories. For instance, the number of rain events with 850-mb winds less than 10 knots (six events) was approximately the same as the number with 850-mb winds equal to or greater than 10 knots (eight events). The category with the lighter wind speed for each pressure level corresponds to "light winds", whereas the other category corresponds to "strong winds". Light winds were closely correlated with highly acidic events. For the period considered here, the average acidity was about 3.4 times greater, when light winds prevailed 24 to 48 hours before a rain event, and about 2.6 times greater, when light winds prevailed up to 24 hours before the rain event. In only one category, 700-mb winds up to 24 hours before rainfall, did light winds tend to decrease acidity.

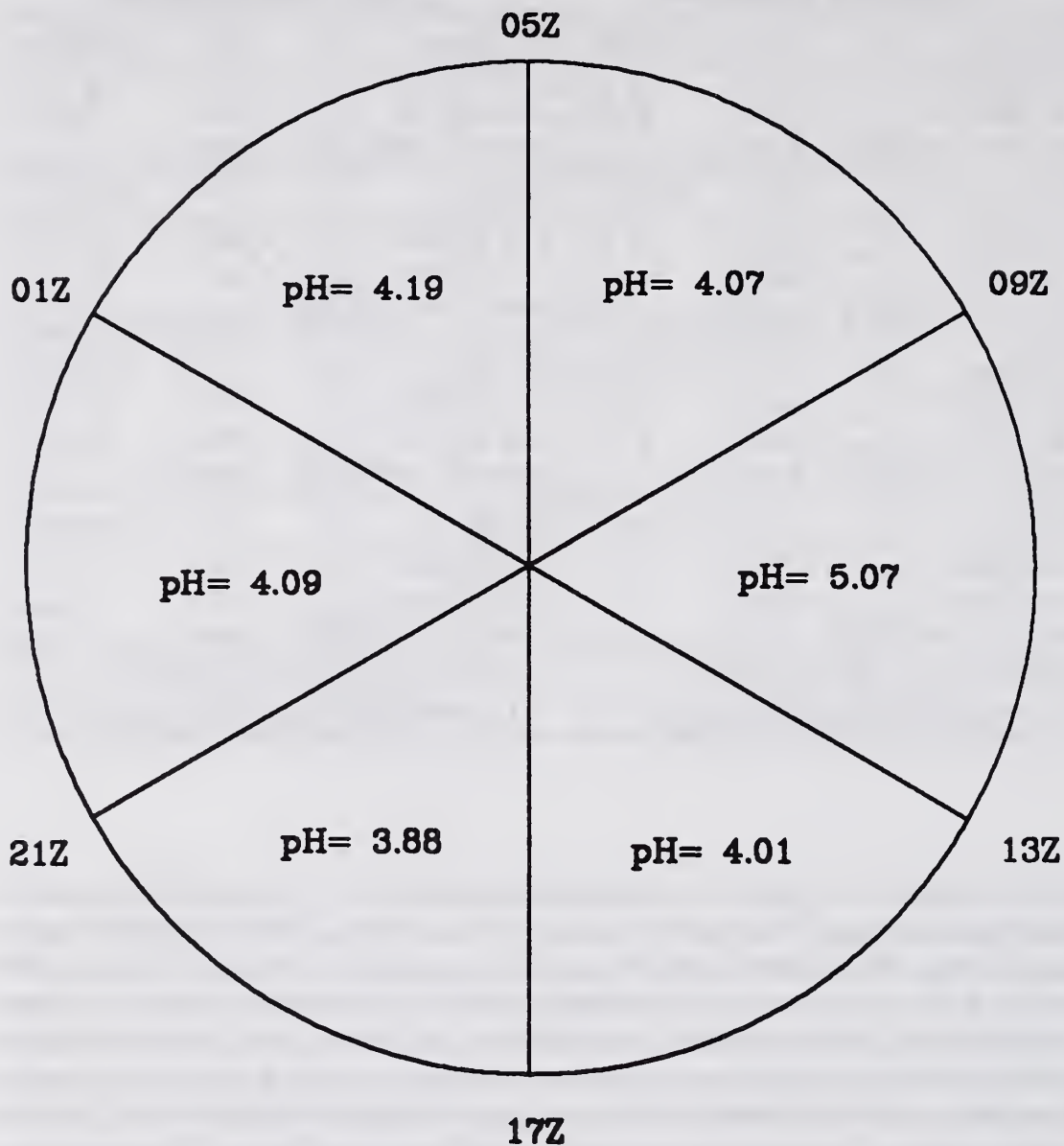
Average pH Values During the Day and Night

FIGURE 5. Effect of time of rainfall on acidity.

Rainfall amount. Another factor closely correlated with acidity was the amount of rainfall in an event. Figure 2 shows the average acidity, expressed in terms of $[H^+]$ for each type of rain event, categorized by amount of rainfall. Six events in the range 0.01 to 0.05 in. are listed as "light", and six in the range from 0.05 to 0.15 in. are listed as "moderate", while the six heaviest rainfalls, all in the range 0.15 to 2.00 in., are termed "heavy". The average pH was 3.9 for heavy rains, 4.3 for moderate rains, and 4.7 for light rains. A high degree of variation in acidity occurred, however, within this trend. The acid concentration varied from the least acidic event to the most acidic event for each rainfall type by a factor of 230 for light rains. In spite of this large variation, the average acidity of heavy rains was significantly greater than that of moderate rains (3 times as much).

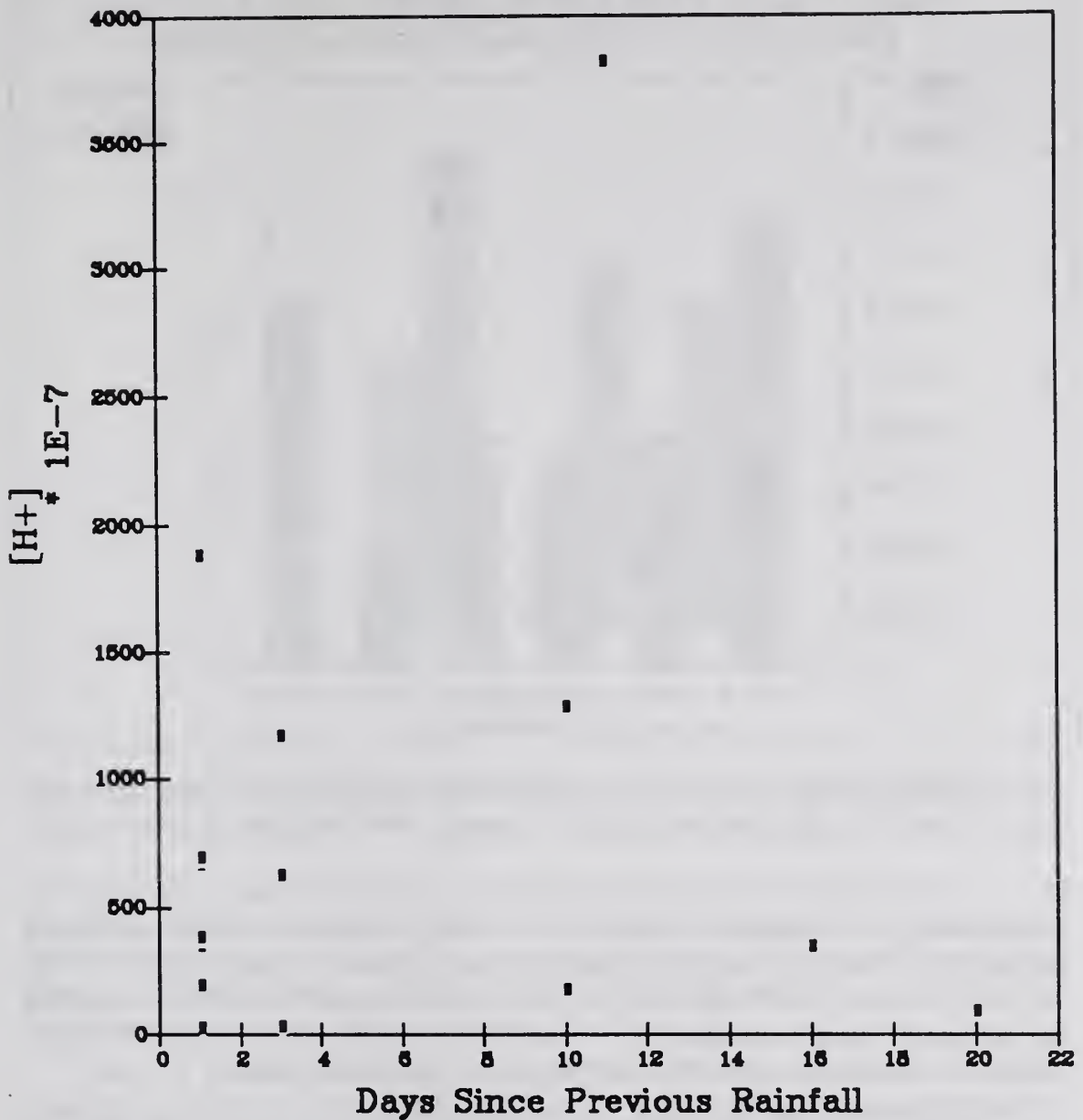
[H+] vs. Days Since Previous Rainfall

FIGURE 6. Effect of frequency of rainfall (day since previous rain) on acidity (all 18 events are shown, but some are clustered due to the scale used).

When the rainfall is very light, contamination of small samples through reaction with the walls of the collector is a concern. Experience (and a few simple laboratory checks) indicates that this effect should result in a decrease in the acidity of the sample, i.e., in an increase in its pH. It might also be anticipated that the longer the sample remained in the collector, the greater the decrease in acidity. However, no indication of this effect was found, as the two least acidic samples from the entire collection period (both from light rainfall) were analyzed within short times after the end of rainfall.

Rainfall rate and duration. Because the amount of rainfall in an event is determined by the rate and the length of rainfall, these two variables were analyzed to determine whether a similar correlation existed between their values and rainfall acidity. Average rates of rainfall were determined by dividing the

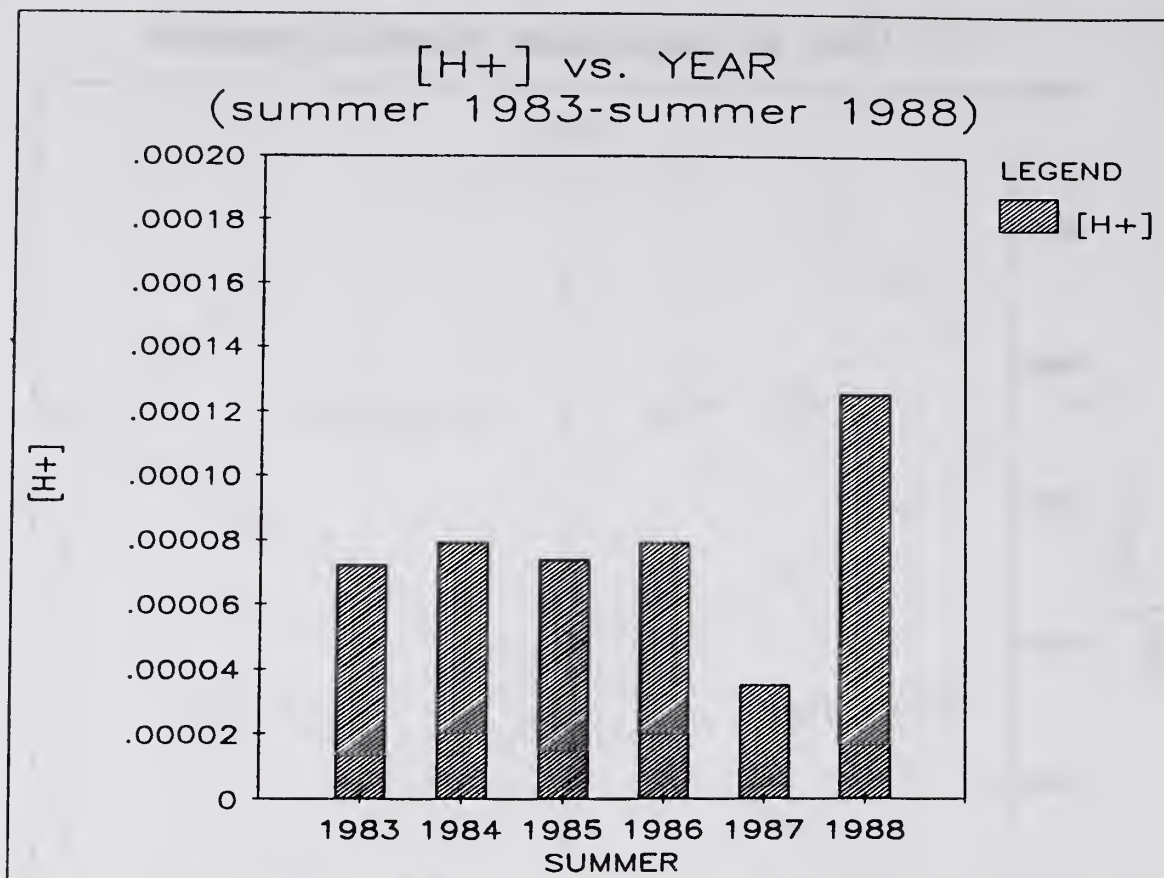


FIGURE 7. May to July averages of rainfall acidity at the collection site (six years; data for 1983 through 1987 from NADP records; 1988 data from current study).

total amount of rainwater collected by the total duration of the contributing rainfall. The resulting values are shown in the rightmost column of Table 1. For 14 of the 18 cases, there was only one rain event during the 24-hour sampling period; using a single average rate for the four days with two or more rain events in the 24 hours did not greatly effect the results presented here.

Figure 3 shows the relationship between the rate of rainfall and acidity. The rates were grouped into categories according to intensity. Five rainfall rates in the range 0.01 to 0.04 inches/hour are characterized as "slow", seven in the range from 0.04 to 0.06 inches/hour as "moderate", and six in the range from 0.06 to 0.50 inches/hour as "fast". Although fast rates (pH = 3.9) produced greater acidity than slow rates (pH = 4.2), the lowest acidity was associated not with slow rates but with moderate rates (pH = 4.8). Thus, no clear correlation is apparent between the rate of rainfall and rainfall acidity.

A much clearer correlation exists between the duration of rainfall and acidity (Figure 4). Once again the data were grouped into 3 approximately equal-size categories according to size. The six shortest rainfalls in the range (0.25 to 1.25 hours) are characterized as "short", the seven longest rainfalls in the range (4 to 4.5 hours) as "long", and the five remaining as "moderate". A pattern emerges that is similar to that between rainfall amount and rainfall acidity (Figure 2). Long rainfalls (pH = 3.9) tended to have more acidity than moderate rainfalls (pH = 4.3), which in turn tended to have greater hydrogen ion activity than short rainfalls (pH = 5.0). On the average, long rains were two times more acidic than

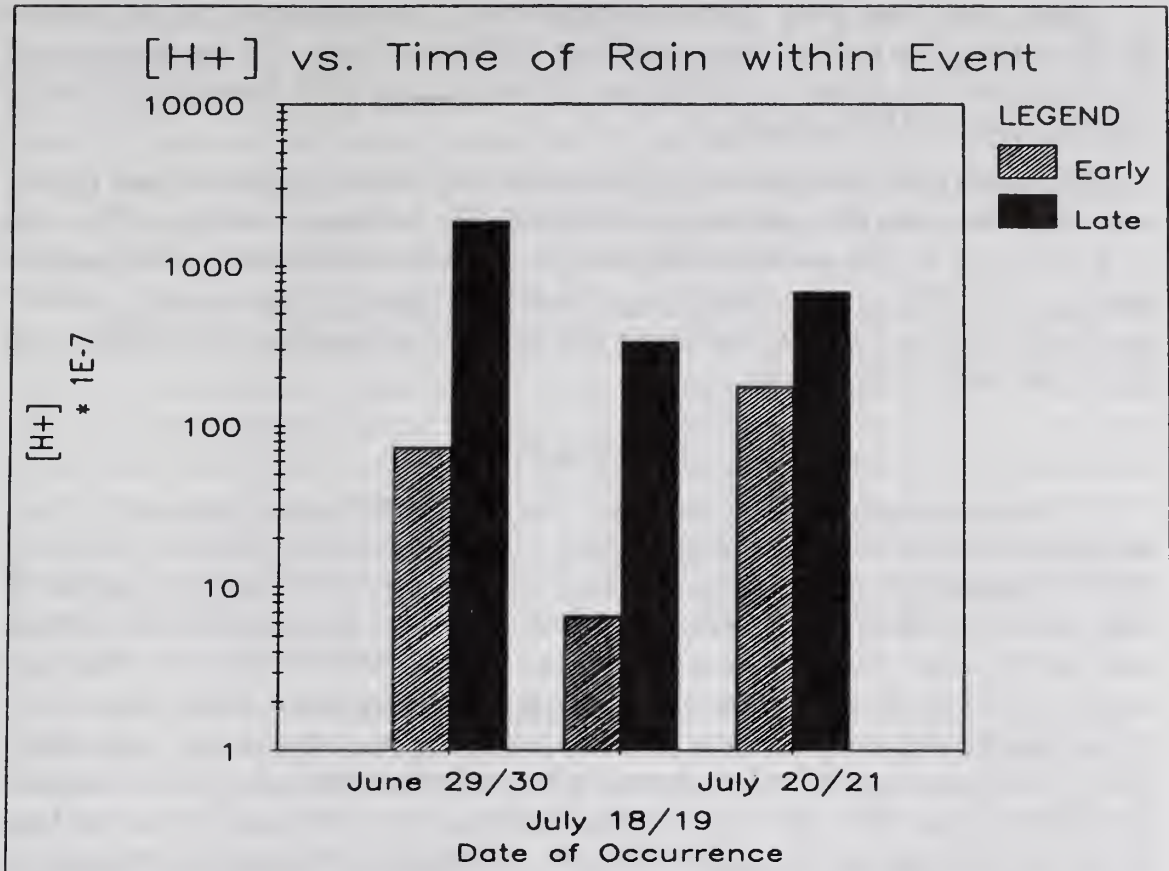


FIGURE 8. Effect of time of rainfall within a rain event on acidity (3 cases).

moderate rains and 14 times more acidic than short rains. Once again, a large amount of variation occurred within this trend. The hydrogen ion activity varied from the least acidic event to the most acidic event for each rainfall length by a factor of 22 for long rains, a factor of 16 for moderate rains, and a factor of 120 for short rains.

Time of rainfall. The time of rainfall also affected the acidity. Figure 5 represents a 24-hour clock partitioned into six four-hour intervals, starting at 5Z (midnight local time). The slices represent time intervals within which a rain event occurred. If an event spanned two or more time intervals, the pH of that event was averaged into each interval within which it occurred. The most acidic rainfalls occurred between 17Z - 21Z (noon - 4 p.m. local time), and the least acidic rainfalls occurred between 09Z - 13Z (4 a.m. - 8 a.m. local time). If one were to analyze daytime and nighttime rainfalls from the data, the approximate time intervals would be 01Z - 13Z (8 a.m. - 8 p.m.) and 13Z - 01Z (8 p.m. - 8 a.m.), respectively. The average daytime pH was 4.1, approximately 2 times as acidic as the average nighttime pH of 4.4.

Effect of preceding dry spells. Although intuitively one might expect more acidic rains following long dry spells due to the accumulation of pollutants in the atmosphere, this expectation is not supported by the data. Figure 6 shows the correlation of acidity with the number of days since the previous rain. This summer's drought provided a wide range of this independent variable (1 day to 20 days), but no correlation is apparent. The correlation coefficient (r) is only 0.14, indicating that acidity was probably independent of the time since the previous rainfall.

May, June, and July 1988 in perspective. A comparison of rainfall acidity for these 18 events in May, June, and July 1988 vis-a-vis that for the same period in recent years is presented in Figure 7. The data for 1983 through 1987 were obtained from NADP records.

The acidity of the 1988 summer's rainfall ($\text{pH} = 3.9$) is greater than that of any of the previous five summers and exceeds the five-year average ($\text{pH} = 4.2$) by a factor of 1.8 (80% greater than normal). A trend in summertime acid concentration since 1983 is not evident from the data. Although this summer's rainfall was much more acidic than the 5-year average, last summer's rainfall (1987) was much less acidic ($\text{pH} = 4.5$).

DISCUSSION

Pollution sources as far as 500 miles away can contribute significantly to wet deposition at a location, if the prevailing upper-air winds are favorable (GAO, 1984). Therefore, it is to be expected that when winds aloft are from regions of high sulfur-dioxide-emission prior to a rain event, the resulting rainfall will be more acidic than when these winds are from a region with few sources. The rain events in this study clearly show a tendency to be more acidic when winds aloft were from the regions of the heaviest burning of coal by power plants in the Ohio Valley (southeast, south, and southwest of the collection site) prior to a rain event. In addition, when light winds aloft prevail before a rain event, pollutants are less likely to be diluted by either eddy diffusion or forward stretching (the latter term refers to the dilution of pollutants as they are stretched in the direction of the wind, thereby reducing the concentration of pollutants in the air; Oke, 1978). For this reason, light winds aloft tended to produce highly acidic events.

As was mentioned in Data and Analysis Section, the amount of rainwater collected is a function of both the rate and the duration of rainfall. Since heavy rainfall events tended to be highly acidic, it is therefore reasonable to expect long rain events to be highly acidic as well. However, the question then arises as to why heavy rainfalls tended to be more acidic. A possible explanation stems from the fact that acid compounds are created within a cloud from oxidized sulfate and nitrate particles (SO_4 and NO_3) and their unoxidized gases (SO_2 and NO_3), with the compounds resulting from particles tending to be less acidic. Relatively alkaline solutions (derived from airborne soil particles) are also present and decrease the overall acidity of the cloud. If these particles of relatively higher pH are washed out at the beginning of a rain event, then the latter stages of the event will consist of raindrops which are more acidic. Perhaps, then, it is usual for a certain amount of less acidic rain to fall before the more acidic rain occurs. Thus, light rains and short rains might never reach the point when the more acidic rain begins.

This conjecture is supported by three instances during this study in which rainfall from the same weather system occurred both before and after a collection time. In all three instances, the rainfall at the latter stage of the event was more acidic, usually by large amounts (Figure 8 and Table 1).

Because rain dominated by convection tends to be more acidic than frontal-induced rain (General Accounting Office, 1984), the most acidic events should occur during the period of peak convective activity. The higher acidity during the day and the especially high acidity from 17Z - 21Z (Figure 5) likely reflect this convective influence.

The lack of correlation between acidity and time since the previous rainfall (see Figure 6) is possibly due to the combined effects of dry deposition and atmospheric dust. If there is no rain for an extended period, ultimately dry deposition would be expected to balance emissions of pollutants into the atmosphere; the actual deposition pattern will be determined by the winds. This would suggest that after a sufficient period of time without rain, the first rain would be very acidic, if the atmospheric load of pollutants was large.

However, at the same time the pollutant load is building up, the amount of dust (which in general is alkaline and thus serves to neutralize acidic compounds) in the air increases as the surface dries out; lofting of dust depends on wind speeds and the extent of agricultural activity. Thus, when rain does finally occur after an extended dry period, the resulting acidity will depend on the relative proportion of pollutants and dust existing at that time. In a situation with significant dust in the air, a portion of the acidity of the rain would be neutralized, off-setting the increased load of pollutants in the air. This balance process was probably especially important in the drought of 1988 (37% of normal rainfall) with its numerous long periods of extreme dryness.

Why then was the rainfall in May through June 1988 still so much more acidic than that of the same periods in previous years (Figure 7)? A likely explanation would appear to be related to the fact that the extreme heat and dryness in 1988 was produced by an upper-air wind pattern that was dominated by an unusually persistent light, southerly flow that established itself in early May. Such a pattern would have strongly favored transport of an increased amount of pollutants from high SO₂-emitting regions to the study site and at the same time would have reduced dilution of pollutants by forward stretching or eddy diffusion while enroute. In addition, the persistent heat (29 days above 90° vis a vis normal of 14 days) and dryness of this period reflect the fact that those few fronts that did move through the area were weak and generally dry and did not lead to a significant change in the airmass over the droughty region. Most of the rain that did occur was produced by convective showers forming within the airmass and was, therefore, more acidic.

SUMMARY

The May, June, and July 1988 collection period was unusual in that only 37% of normal rainfall occurred, and the acid concentration greatly exceeded the 5-year summer average (1988 average pH = 3.90; 5-year average pH = 4.15). The 18 rain samples collected during the period indicate very acidic rainfall (50 times as acidic as naturally-occurring rain), although a great variation of acidity occurred (lowest pH = 3.42, highest pH = 6.53). When winds aloft were from known regions of high SO₂ emissions (southwest, south, and southeast of collection site) prior to a rain event, the resulting acid concentration of the rain was large compared with instances when winds were from other directions. In addition, wind speeds also affected the acidity, as light winds aloft produced higher acid concentrations in rainfall than strong winds. Heavy rains and long-lasting rains also produced relatively high acid concentrations, as did rainfall that occurred later in an event. The rate of rainfall, however, did not seem to affect acidity. Rain which occurred during the day produced higher acidity than nocturnal rain, with the most acidic rainfalls occurring between 17Z - 21Z. The time elapsed since the previous rainfall did not affect a rain event's acidity.

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