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METEOROLOGICAL AND CHEMICAL FACTORS CONTROLLING THE COMPOSITION OF PRECIPITATION IN EASTERN NORTH AMERICA

by
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ABSTRACT

Precipitation in eastern North America is characterized by high concentrations of free acidity and sulfate that are generally attributed to anthropogenic air pollution. The relatively long record of precipitation chemistry measurements at the Penn State MAP3S site is used to analyze the seasonal and interannual variability of precipitation composition in terms of specific mechanisms of atmospheric transport and chemical transformation. The interrelationships of the chemical variables in the precipitation record and in recent air measurements clearly link the precipitation acidity with the wet deposition of sulfate derived from the in–cloud oxidation of sulfur dioxide. High–deposition events are shown through meteorological trajectory analyses to be associated with moist air from the Gulf of Mexico that passes through the upper midwestern parts of the United States. The main chemical factor controlling the deposition of sulfate appears to be the availability of strong oxidants for transforming dissolved sulfur dioxide into aqueous sulfate. Excess sulfur dioxide is expected to exit the storm systems at high altitudes and experience truly long–range transport. This interpretation of the data gives confidence that episodes of high sulfate deposition will occur even after sulfur dioxide emissions have been reduced substantially.

1. INTRODUCTION

Precipitation in the eastern portions of North America is known for its large content of free acidity (Barrie and Hales, 1984; Dana and Easter, 1987). Even though it has been recognized for some time that the precursors of this acidity have anthropogenic origins (Likens, 1976), many uncertainties remain over the specific processes responsible for transporting and transforming the original emissions into the anionic compounds found in the precipitation of the region. As might be expected, both meteorological and chemical phenomena have been found to be important for the formation of the acidity in precipitation (Clark et al., 1987; Fung et al., 1991).

A fundamental issue still facing the scientific community is the response of acidic deposition to changes in the rates of precursor emissions. It is generally acknowledged that in–cloud processes cause the deposition of sulfate, the dominant anion, to be nonlinearly related to the amount of sulfur dioxide in the air (Clark et al., 1987; Easter and Luecken, 1988), although this view is not universally held (Tremblay, 1987). Analysis of precipitation chemistry data over the eastern United States (Butler and Likens, 1991) shows a general relationship between the concentrations of sulfate and hydronium in precipitation and the emissions of sulfur dioxide at most sites, but not at all. Data from the Penn State atmospheric

chemistry site, in particular, were found to exhibit no discernible relationship to the declining precursor emissions rates. It is the purpose of this paper to present some of the Penn State data in greater detail and to identify the likely meteorological and chemical reasons for the persistently large rainfall acidity in this region.

2. DATA

The data used for this study were derived from the routine collection of precipitation at the Penn State atmospheric chemistry research site in rural central Pennsylvania. The collection and analysis protocol used was that set up under the Multistate Atmospheric Power Production Pollution Study (MAP3S; MacCracken, 1978; MAP3S/RAINE, 1982). From the start of sampling in 1976 until 31 January 1988, precipitation was collected after every meteorological event. The minimum interval between collections was 24 hours, whereas the interval could extend for several days during persistent periods of rain. Since 1 February 1988 the sampling interval has been held constant at 24 hours, so some meteorological events are now resolved into several samples. The results presented here thus stem from a mixture of event and daily samples, although the term "event" will be used generically to represent the entire data set. The precipitation chemistry data are all available in the data base of the Acidic Deposition System (ADS; Watson and Olsen, 1984).

For various purposes the precipitation chemistry data have been aggregated into distinct time periods. When concentration information is needed, a weighted average has been formed based on the amount of precipitation sampled. Deposition amounts have been calculated as the product of the event (or daily) concentrations and the precipitation depth, using the associated Belfort rain gage data, and then summed over the particular period of interest.

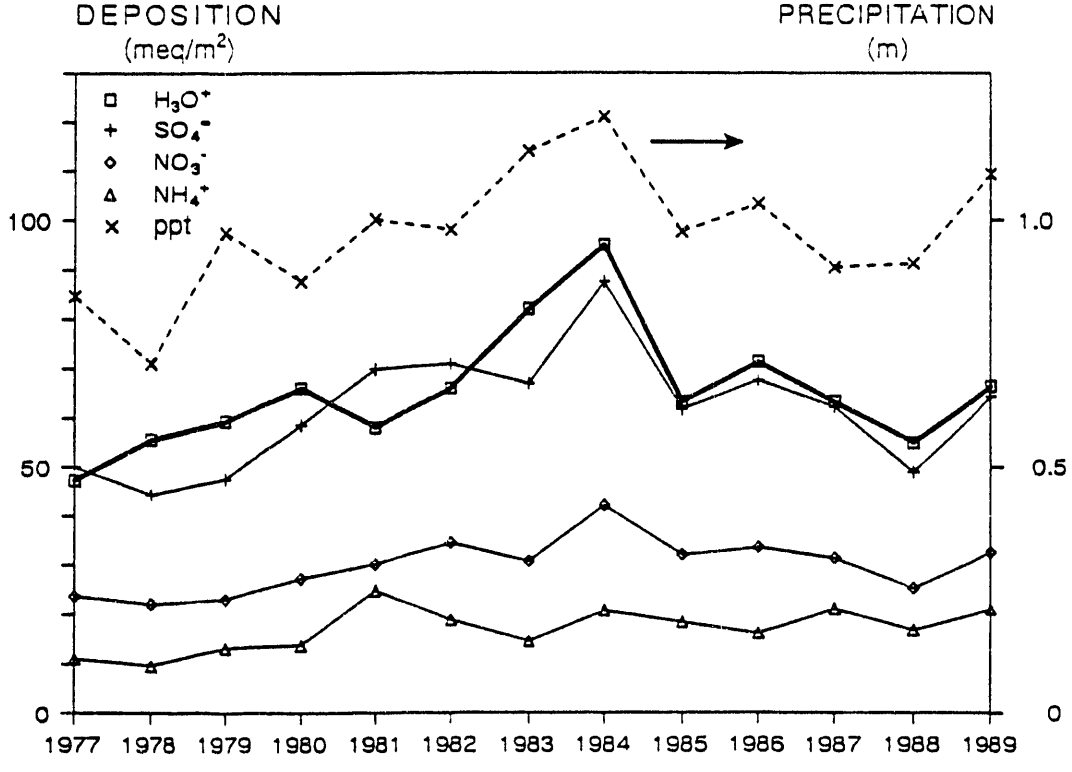

FIGURE 1. Annual deposition of ions and precipitation at Penn State MAP3S site.

Annual deposition amounts for each full year of the available MAP3S record are shown in Figure 1 along with the annual precipitation depths. The bold line shows the year to year variations in the deposition of the hydronium ion. To a reasonable first approximation, the interannual variability reflects that of the sulfate deposition, much less so those of the nitrate or ammonium ions. Much of the variability in chemical deposition is also driven by variations in annual precipitation. The annually averaged concentration data indeed show relatively little year to year changes and no significant long–term trend, as suggested by other analyses (Dana and Easter, 1987; Butler and Likens, 1991). The year 1984 was however relatively wet in central Pennsylvania and yielded precipitation of particularly high ionic concentrations.

As the period of averaging is reduced, the natural variability of acidic deposition increases. Figure 2 shows these same data broken apart by season. The depositions of both hydronium (Figure 2a) and sulfate (Figure 2b) are seen to vary greatly from season to season and even from year to year within any given period. Invariably, summer is the season of maximum precipitation acidity in eastern North America. The relatively large depositions of acidity and sulfate that showed up in the 1984 annual averages can now be identified with an unusually wet and polluted summer period.

When the precipitation chemistry data are averaged on a monthly basis, as in Figure 3, the climatological maximum in chemical deposition is clearly seen to occur in the summer months of June, July, and August. Some of the intra–annual variability in deposition is driven by the seasonally dependent precipitation amounts, but certainly not all of it. Likely chemical mechanisms for the annual signal will be discussed in the next section. As we break out the contributions to the monthly averaged sulfate deposition by the individual months of data in the record, as shown in Figure 4, we again find very large variations. Particularly revealing is the large deviation from the climatological mean in August of 1984. The fact that the deposition of sulfate in August 1984 was almost three times the long–term average is clearly the basis for the relatively large 1984 annual depositions of sulfate and acidity that were shown earlier (Figure 1).

3. ATMOSPHERIC PROCESSES

The relatively long MAP3S data record contains a wealth of information that helps us identify and understand the atmospheric processes that account for the appearance of the diverse chemicals in precipitation. A particular strength of the MAP3S protocol, namely the high temporal resolution of the sampling and its identification with individual meteorological events, can be used here.

The very large wet deposition of sulfate in August 1984, for instance, can be traced back to the occurrence of a number of unusually high–deposition events, combinations of large storm precipitation amounts and high sulfate concentrations, despite the natural tendency for dilution of solutes in large quantities of water. Detailed analyses of the meteorological situations in these cases have shown the prevailing air flow to have origins in the Gulf of Mexico during periods of high pressure in the southeastern United States. Figure 5 shows the results of an air trajectory analysis (Heffter, 1983) that was performed for one of the high–deposition cases. The warm, moist air that spawned the storm on 9 August 1984 in central Pennsylvania had passed along the emissions–rich Ohio River Valley the previous two days. The meteorological stage was thus set for funneling both moisture and pollutants into the region of the receptor site.

The composition of the rain that fell during this particular high–deposition event yields insight into the chemical factors responsible for the acidity of the rain. The precipitation–weighted mean pH of the rain was 3.8, a value that can be accounted

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