ANALYSIS OF SOME ASPECTS OF THE SURE AIRCRAFT SULFATE DATA

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Abstract—Aircraft data collected during the first five intensive and first episode periods of the Sulfate Regional Experiment (SURE) were analyzed through the lowest 3 km of the atmosphere to investigate possible long-range transport and transformation of sulfur pollutants. Results indicate average sulfate concentrations in the lowest 3 km over several stations generally higher in summer than winter, higher in afternoon than morning, and higher in the lower half of the layer than the upper half. Trajectory analyses for episode and non-episode periods in July 1978 show sulfur pollutants emitted in the Ohio River Valley possibly responsible for high sulfate concentrations in rural Pennsylvania and New York.

Key word index: Long range transport and transformation, sulfate, Sulfate Regional Experiment, sulfur dioxide, vertical sulfate climatology, trajectory analysis.

INTRODUCTION

Sulfate concentrations have been estimated as $0.8 \mu\text{g m}^{-3}$ over the North Atlantic (Prahm and Christensen, 1979), $1 \mu\text{g m}^{-3}$ over the United States (Altshuler, 1976), and $1.5 \mu\text{g m}^{-3}$ over Canada (Chung, 1978). However, urban sulfate values are as high as $83 \mu\text{g m}^{-3}$ under adverse meteorological conditions (Tong *et al.*, 1976). Formation of sulfate aerosols by oxidation of atmospheric sulfur is temperature- and moisture-dependent and thus surface sulfate concentrations are higher in summer than winter, even though winter sources of atmospheric sulfur are stronger.

Afternoon sulfate values between 0.3 and 1.5 km above Philo, Ohio, as measured by the Sulfate Regional Experiment (SURE), were consistently higher than morning values during August 1977. According to Mueller *et al.* (1979), this result could be caused by "differences in day-night (insolation-sensitive) formation rates." This also explains why almost no difference is seen between their corresponding afternoon and morning January values, i.e. low insolation period values. Little data exists on vertical differences in sulfate concentrations on the regional scale, but observations at Rockport, Indiana during one morning and afternoon in August 1977 showed lower concentrations in the layer from 1.5 to 3.0 km than in the layer from 0.3 to 1.5 km (Mueller *et al.*, 1979).

Afternoon aircraft observations taken 100 km east of New York City (NYC) during January 1978 by

Michael *et al.* (1979) as part of the Multistate Air Pollution Power Production Study (MAP3S) showed sulfate values within the mixed layer from one to two orders of magnitude higher than those in the layers above. However, the fraction of sulfur in the form of sulfate was three to eight times larger above the mixed layer than within it.

Although sulfur dioxide levels in East Coast urban centers have generally decreased because of the change to low sulfur fuels and relocation of power plants to the Ohio River Valley, sulfate levels in these areas did not decrease accordingly (Altshuler, 1976). This 'sulfate anomaly' is believed to be due to long-range transport of sulfur dioxide accompanied by its simultaneous chemical transformation to sulfate. Long-range transport is believed to be enhanced by the tall stacks used with the new Ohio River Valley sources to reduce local sulfur dioxide concentrations.

Previous observational studies indicate long-range transport of sulfate and its precursors related to high local ambient sulfate concentrations, e.g. Wolff (1979) found 75 per cent of the sulfate in New York State (NYS) not produced within the state. Sulfur dioxide roses constructed by Zeeduk and Velds (1973) showed concentrations in the Netherlands dependent on wind direction. That study, and one by den Tonkelaar (1978) using a surface monitoring network, showed the industrial Ruhr Valley producing high sulfate concentrations in the Netherlands. Visual imagery from the SMS1 satellite was used by Lyons *et al.* (1978) and Parmenter (1977) to follow pollutant clouds from the midwest to the northeastern United States.

Analyses by Samson (1978, 1980a) and Galvin *et al.* (1978) showed sulfate and ozone transport over long distances producing high concentrations in rural NYS. Similar analysis by Milan and Chung (1977) showed

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high sulfate concentrations in southern Quebec resulting from long-range transport from the south. In addition, the Diagnostic Atmospheric Cross-Section Experiment (DASCE-1) of MAP3S showed sulfate transport taking place on isentropic surfaces (MacCracken, 1979).

The SURE backwards trajectory model described by Hidy *et al.* (1976) used 2000 foot (~ 610 m) winds to construct 24-h long-range transport trajectories. Similar 72-h trajectories were constructed by Miller *et al.* (1978), Samson (1980a, b, c), and Gotaas (1982). The four level (surface to 1600 m) numerical diagnostic trajectory model of Hefter and Taylor (1975) was used to construct similar long-range transport trajectories over the eastern U.S. by Lioy *et al.* (1980).

Preliminary results from SURE (Lavery *et al.*, 1979) show two different synoptic patterns associated with long-range transport and high rural sulfate 'episode' concentrations. The first, a 'stagnation' situation, involves a slow moving anticyclone which collects primary pollutants from the industrial areas of the Ohio River Valley and transports them slowly eastward. The second involves 'channeling,' in which a duct is established between the Appalachian Mountains and a weak cold front oriented west-southwest to east-northeast and travelling southeastward.

In the current study, sulfate and sulfur dioxide data collected by research aircraft as part of the SURE experiment are used to prepare preliminary concentration climatologies through the lowest 3 km of the atmosphere over the northeastern United States. Radiosonde data are used to construct long-range 850-mb isobaric trajectories for one low sulfate concentration period and one sulfate episode period during July 1978.

ANALYSIS

Instrumented aircraft were flown during the first five SURE routine 'intensive' observational periods and one high concentration 'episode' period (designation by SURE investigators based on surface concentrations) during 1977 and 1978 (Table 1). A Beechcraft Queen Air was flown by Meteorology Research Inc. (MRI) and a Piper Navajo was flown by Research Triangle Institute (RTI). The aircraft measured particulate and gaseous pollutant concentrations, as well as temperature, dew point, turbulent intensity, sky cover and visibility.

Both planes were equipped with a Meloy 285 flame photometric sulfur dioxide device. In addition, 47-mm coated filters, manufactured by Brookhaven National Laboratory, were also used to measure sulfur dioxide starting in the RTI aircraft in January 1978 and the MRI aircraft in April 1978. The Navajo aircraft used the same filters for measuring respirable sulfur particulates, whereas the Beechcraft used Environmental Research and Technology instrumentation for this purpose. Details of instrumentation, as well as preliminary listings of unvalidated data, are in Keifer (1979).

During intensive periods, each aircraft collected data in the vicinity of the SURE stations listed in Table 1. During the episode period, the aircraft did not remain near a single station, but flew traverses between the two stations listed in the Table. All sites listed in Table 1 are located in Fig. 1.

On each sampling day during an intensive period, both morning "a.m." and afternoon "p.m." flights were flown by each aircraft. The early flight was near sunrise or midmorning, while the later flight was midafternoon or early to late evening. Each flight consisted of a vertical spiral after takeoff, a horizontal traverse to the flight midpoint, a second vertical spiral, a second horizontal traverse, and a final vertical spiral. A single filter was used for sulfate and sulfur dioxide in the lower halves of all three spirals (near surface to 1500 m), and a different one for the upper halves of all three spirals (1500–3000 m). On days during the episode period, each plane flew from one airport to the other in the morning and then back in the afternoon. Filters were changed at the midpoint of each flight, yielding two observational values per flight.

To produce a 'vertical sulfate climatology' for the SURE

Fig. 1. Map of SURE region showing locations of observational sites.

region, concentration data from both sites on each sampling day (Table 1) during a particular intensive period were used to compute average values of sulfate content in a unit-area column for the morning lower layer, morning upper layer, afternoon lower layer, and afternoon upper layer. Concentration values were multiplied by column depth to produce sulfate content values in $\mu\text{g m}^{-2}$. Since data were collected at Philo during four of the five intensive periods, the same averaging procedure was used to compute a separate climatology for this station. A similar procedure had been used by Mueller *et al.* (1979) for Philo data from August 1977 to January 1978.

The maximum recorded sulfate value in the upper layer ($22.4 \mu\text{g m}^{-3}$), from the morning of 16 July 1978, at Philo, appeared excessive when compared with other upper layer data. Thus, an average of morning observations taken immediately before and after this day ($1.6 \mu\text{g m}^{-3}$) was used in its place. A similar procedure replaced a sulfur dioxide reading of $90.0 \mu\text{g m}^{-3}$ from the second half of the 20 July morning flight from Philo to Rockport with one of $35.0 \mu\text{g m}^{-3}$. It is possible that the first replaced value was due to a point source plume in the sounding. This also could be true for the second replaced value if a significant portion of the traverse was within a plume. Plume contamination is always a potential problem in regional scale problems. Use of the two questionable original data would result in strengthening conclusions reached below.

To investigate the possible role of long-range transport in producing high sulfate values observed during the episode period, backward trajectories ending at Albany, New York were constructed at 12-h intervals. Rawinsonde winds from the 850 mb level from the episode and intensive periods of July 1978 were used as input to the SURE trajectory model (Hidy *et al.*, 1976) to construct 48- to 72-h isobaric trajectories.

RESULTS

(a) Sulfate climatologies

The climatology of total sulfate from near surface to 3 km at Philo (Fig. 2) shows summer maxima and winter minima during both a.m. and p.m. periods, as is also true for the lower layer data. However, this is not true for the upper layer data, as a.m. values during the second Summer are less than corresponding spring values and p.m. winter values are greater than corresponding second summer values. In addition, only in this last case are upper values greater than corresponding lower values. Finally, all p.m. values (i.e. total, upper and lower) are greater than corresponding a.m. values during all seasons.

The preliminary Philo results of Mueller *et al.* (1979) show seasonal and diurnal patterns similar to the above results, even though they only used some of the Summer 1977 and Winter 1978 data from the lower layer and no upper layer values. In particular, they found higher summer than winter values during both a.m. and p.m. periods and higher p.m. than a.m. values in summer, but approximately equal values in winter. Finally, their one day of data from Rockport showed smaller lower-level values during both time periods. The general decrease of sulfate with altitude was also supported by their 24-h-averaged surface sulfate values, which were about three to four times greater than average a.m. and p.m. lower layer readings.

Fig. 2. Climatology of total sulfate in column of air to 3 km at Philo, Ohio, with shaded regions indicating values in upper half of layer.

The 'regional' climatology (Fig. 3) has some features which are consistent with the Philo climatology, however, there are significant differences. For example, in all seasons a.m. and p.m. upper layer values are less than corresponding lower layer values for both Philo and the region. However, summer maxima and winter minima only appear in the total p.m. values, but not the total a.m. values. Finally, p.m. values are not consistently greater than a.m. values for either the total column or its upper and lower layers. Thus, Philo is not always representative of the entire Northeast.

(b) Trajectory case studies

All 12 trajectories computed during the 11–14 July 1978 non-episode period were only 48 h long because of the relatively high wind speeds during the period. In addition, only one (or possibly two) trajectories passed through high emissions areas around the Ohio River Valley (Figs 4a–c). The four trajectories terminating from 1900 EST on the 11th to 0700 EST on the 13th originated in northern or central Ontario (Fig. 4a), far north of the Valley. These later trajectories reflect the presence of a trough at 850 mb.

The more zonal orientation of trajectories on the following two days (Fig. 4b) reflect entrance of a broad short-wave ridge at 850 mb. Although these trajectories are nearer to the Ohio River Valley, only the one terminating at 0700 EST on the 15th actually passed close to it.

Continuation of this synoptic pattern is reflected in the two earliest trajectories illustrated (Fig. 4c) and in

the first third of the trajectory arriving at Albany at 1900 EST on the 16th. However, movement of a closed low at 850 mb to the East Coast during early morning hours on the 15th is reflected in the change to a meridional orientation in the latter two-thirds of this trajectory and in the almost closed loop of the following one.

Trajectories computed for the high sulfate 'episode' period during the third week of July 1978 (Figs 5a and b) were longer than those of the intensive period. They ranged from 48 to 72 h due to the generally lower wind speeds during this period, as compared with those during the intensive period. Four, and possibly five, of the eight trajectories constructed for this episode period passed through high emissions areas around the Ohio River Valley.

The earliest trajectory (Fig. 5a) reflects replacement of the ridge of the 18th by a trough on the 19th, as this trajectory and the second half of the next one (0700 EST on the 19th) did not pass through the heavy emissions area. The final two trajectories of Fig. 5(a), and the earliest one of Fig. 5(b) (i.e. arrival at 1900 EST on the 20th), did slowly pass over the heavy emissions area as the trough moved through. The next trajectory passed north of the emissions area, but that at 1900 EST on the 21st (Fig. 5b) again passed over it. The final trajectory, which passed north of the area, is quite similar to that 24 h earlier. The presence of a ridge which moved into the area on the 21st is seen in these final three trajectories.

As concentration data from the intensive period

Fig. 3. Same as Fig. 3, but for entire SURE region.

consisted of vertical spirals from the surface to 3 km, and data from the episode period consisted of 300 km long horizontal traverses at elevations near 1 km, it is difficult to compare the two data sets. However, data from the intensive period (12 to 16 July) in the lower layer (i.e. 0–1500 km) can be compared with those from the episode period (19–21 July).

Results (Fig. 6) show average concentrations of both pollutants higher during the episode than during the intensive period, with sulfur dioxide levels showing the greater percentage increase. Thus, the general 'sulfate greater than sulfur dioxide' pattern of the intensive period reversed throughout the episode.

Two caveats should be kept in mind when viewing the above results. First, traverse data from the MRI aircraft were taken along paths from Scranton to Galloo on the 19th and 20th and from Scranton to Montague on the 21st (Table 1). Second, each morning and evening data point for a given day represents either the average of two traverses (for episode period) or one sounding each from two sites (for intensive period).

Average concentrations of both pollutants were higher at Philo than at Scranton during the intensive period (Table 2). This is expected given the trajectories illustrated in Figs. 4(a)–(c), as Scranton is only in-

frequently downwind of the large sulfur dioxide sources around Philo. Average sulfur dioxide concentrations were also higher along Rockport to Philo traverses than along Galloo or Montague to Scranton traverses during the episode period (Table 2), when the trajectories of Figs 5(a) and (b) show Scranton frequently downwind of the large sulfur dioxide sources around Philo. However, sulfate concentrations were higher along Scranton traverses during this period, thus providing evidence for sulfur dioxide to sulfate transformation in conjunction with the long-range transport implied by the trajectory analysis.

The position of a surface cold front during the 'episode' was similar to that which produced long-range transport of the channeling type detailed by Lavery *et al.* (1979). In addition, high temperatures and humidities, as well as low wind speeds at the surface and 850 mb during the episode, favored a more rapid and complete conversion of sulfur dioxide to sulfate during movement to Scranton. Additional evidence for transformation is found in the higher *R*-values (ratio of sulfur content of sulfate to sulfur content of sulfur dioxide plus sulfate) along Scranton traverses than along Philo traverses during the episode. This is reversed from conditions during the intensive period (Table 2).

Table 1. Aircraft data collection protocol during SURE intensive periods and episode period (shown by ‡) collected by aircraft

Dates	RTI Aircraft	Sites MRI Aircraft
1977		
1, 3, 5, 7, 9, 11 August	Philo	Rockport
19, 21, 23, 25, 29, 31 November	Scranton	Rockport
1978		
22, 24, 27–31 January	Lewisburg	Philo
12, 14–16, 18, 20–22, 24 April	Research Triangle	Philo
12, 14, 16 July	Philo	Scranton
‡19–21 July	Philo to Rockport	Galloo to Scranton (on 19 and 20 July) and Galloo to Montague (on July 21)

Table 2. Concentration data for July 1978 intensive and episode periods, where *R* is the ratio of sulfur content of sulfate to total sulfur content (sulfur dioxide and sulfate)

Sites	Sulfate ($\mu\text{g m}^{-3}$)		Sulfur dioxide ($\mu\text{g m}^{-3}$)		<i>R</i> (Per cent)
	Range	Average	Range	Average	
Intensive (soundings)*					
Philo	5.02–27.8	15.9	1.6–32.3	16.3	36.9
Scranton	1.0–13.0	7.0	6.6–15.8	10.0	29.6
Episode (traverses)†					
Rockport to Philo	11.4–45.9	26.8	11.4–55.5	38.6	29.4
Galloo to Montague or Scranton	10.1–46.8	31.9	11.6–56.7	28.7	40.0

*Averages of two soundings each on three days.

†Averages of two segments each for two traverses each on three days.

CONCLUSIONS

Sulfur dioxide and sulfate concentration data collected by research aircraft in the lowest 3 km of the atmosphere over the northeastern United States during five seasons from Summer 1977 to Summer 1978 as part of the SURE experiment have been analyzed. Rawinsonde data from July 1978 were used to construct 850 mb isobaric long range trajectories during a low sulfate concentration intensive period and a high sulfate concentration episode.

Results showed seasonally averaged sulfate values in the column from near surface to 3 km at Philo to be greatest in summer and lowest in winter during both morning and afternoon hours. This pattern was also true in the lower half of the layer, but not the upper half. All but one of the 10 seasonal averages (five morning and five evening) from the lower layer were greater than

Fig. 4.(a)-(c) Trajectories ending at Albany, New York, during July 1978 intensive period.

Fig. 5.(a) and (b). Same as Fig. 4, but for July 1978 episode period.

corresponding upper layer values. All afternoon values (i.e. total, upper and lower) were greater than corresponding morning values during all seasons. A climatology for the region showed similar results, except that summer maxima and winter minima did not appear in morning data for the total column, and evening values were not consistently greater than morning values for either the total column or its upper and lower halves.

Only one (or possibly two) of the computed 12 trajectories passing over Albany during the low concentration intensive period of July 1978 first passed over the high emissions area around the Ohio River Valley. However, four (or possibly five) of the eight trajectories passing over Albany during the high concentration episode period did pass over that area.

Daily averaged morning and afternoon concentrations of both sulfur dioxide and sulfate during July 1978 were generally higher during the episode than during the intensive period, with sulfur dioxide levels showing the greater percentage increase. Average concentrations of both pollutants were higher at Philo than Scranton during the intensive period, as expected from the distribution of sulfur dioxide sources and the trajectory analysis.

Average SO_2 concentrations were also higher at Philo than Scranton during the episode, when trajectory analysis showed Scranton frequently downwind of the large sulfur dioxide sources near Philo. However, sulfate concentrations were higher near Scranton than Philo during this period, providing evidence for sulfur dioxide to sulfate transformation in conjunction with the long-range transport implied by the trajectory analysis. The low wind speed, high temperatures, and high humidities at the surface and 850 mb during the episode favored more rapid and complete conversion of sulfur dioxide to sulfate during

movement to the Scranton area. Additional evidence for transformation were the higher ratios of sulfur content of sulfate to sulfur content of sulfur dioxide plus sulfate at Scranton than Philo during the episode, the reverse of that during the intensive period.

The current transport and transformation results are in variance with those of Samson (1980a), who found little correlation between downwind ambient sulfate concentrations and upwind sulfur dioxide emissions. He did, however, find a correlation between upstream stagnation and high downwind sulfate concentration. However, results from Lioy *et al.* (1980) and Tanner and Leaderer (1982) do agree with those of the present study with respect to parcel trajectory and high sulfate concentrations.

Future analyses of this type should utilize concurrent aircraft data collected by multiple aircraft, while future field programs should seek more detailed vertical and temporal resolutions of sulfate concentration. In addition, soundings and traverses for the same and different collection periods should be more comparable.

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Fig. 6. Time variation of area-averaged sulfate and sulfur dioxide concentrations.

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