Case Study of an Unusual Long-Range Sulfur Transport Episode

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ABSTRACT

Observations of extremely high fine particulate sulfur concentrations during early April 1983 in the western United States are linked to a strong cyclone over the midwestern United States. The strong winds around this cyclone circulated polluted midwestern air as far west as the Pacific Coast. A retrograding upper wave pattern was conducive for this polluted air to move southwestward. Both a long-range trajectory analysis and a subjective evaluation of synoptic conditions confirm this hypothesis.

1. Introduction

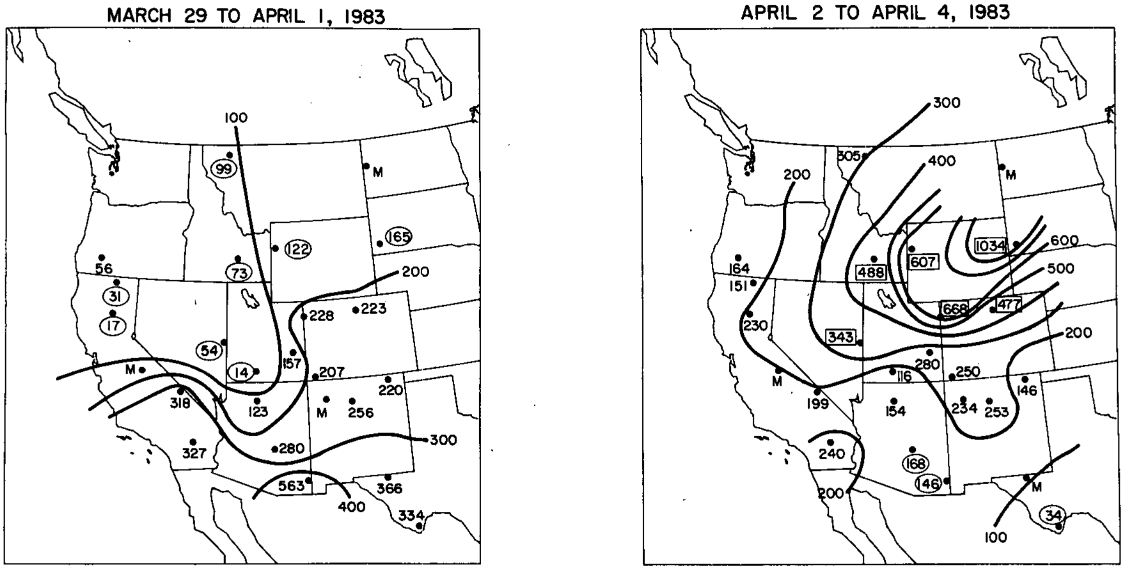
The atmosphere over the United States west of approximately 100°W contains significantly less particulate sulfur than over eastern regions, primarily due to the relatively small number of anthropogenic sources and the arrival of clean air from over the Pacific Ocean. For example, concentrations of fine particulate sulfur (with diameters less than 2.5 μm) are typically four times greater in the East than at sites in the West (Cahill et al., 1985). However, increases in particulate sulfur in the western United States concern the National Park Service (NPS) because of the light (Mie) scattering properties of fine sulfur particles. To help protect the many scenic vistas in the West, the NPS is operating a visibility and particulate data collection network to investigate the effect of particles and their transport upon visibility (Cahill et al., 1985; Malm and Molenar, 1984).

Emissions inventories indicate that the prime anthropogenic sources in the western United States include large urban areas (such as Los Angeles), nonferrous smelters (the largest point sources), and coal-fired power plants (Roth et al., 1985; U.S. Environmental Protection Agency, 1981). The overall pattern of fine sulfur concentrations shows the lowest values in the northwest, north of 40°N and west of 114°W, with concentrations increasing toward the east and south (Cahill et al., 1985). The overall annual cycle of fine sulfur yields highest concentrations during the summer and lowest in the winter at most sites, except for the northern sites, which have a springtime maximum (Cahill et al., 1985). These springtime episodes of high sulfur concentrations in the northern Great Plains were investigated previously (Henmi and Bresch, 1985; Ashbaugh, 1984) and were found to be caused by westward transport of polluted air from the midwestern United States. Ashbaugh et al. (1984) found a springtime maximum in eddy sulfur transport (accomplished by transient cyclones) at sites in the northern Plains. Results from investigations of regional episodes in the intermountain region have indicated source areas in southern California or in the copper smelter region of southern Arizona and New Mexico (Henmi and Bresch, 1985; Ashbaugh et al., 1985; Malm et al., 1984; Pitchford et al., 1981). During the course of our study of long-range sulfur transport into national parks in the western United States, we came across an unusual case during April 1983 in which fine sulfur concentrations were exceedingly large throughout the West yet could not be attributed to transport from local (i.e., western) sources such as southern California or the smelter region. This note describes the meteorological conditions associated with this case.

2. Data and methodology

NPS network particulate data were obtained from the University of California, Davis. These data were collected at approximately 25 national parks, primarily in the West, using a Stacked Filter Unit sampler with two size ranges, 2.5–15 μm and less than 2.5 μm. Samples were 72 h in duration and were analyzed using particle-induced X-ray emission (PIXE) for elemental content. The sites for NPS samplers were chosen to be regionally representative, remote, and away from local particle sources such as urban areas, roads, or chimneys. Because it is difficult to compare these observations directly with weather maps, backward trajectories were computed to help determine air mass source regions.

Air parcel trajectories were calculated using the Air Resources Laboratories Atmospheric Transport and Dispersion (ARL-ATAD) model (Heffter, 1980). Input data consisting of rawinsonde measurements in the lowest 5 km were obtained in NAMER-WINDTEMP



MARCH 29 TO APRIL 1, 1983

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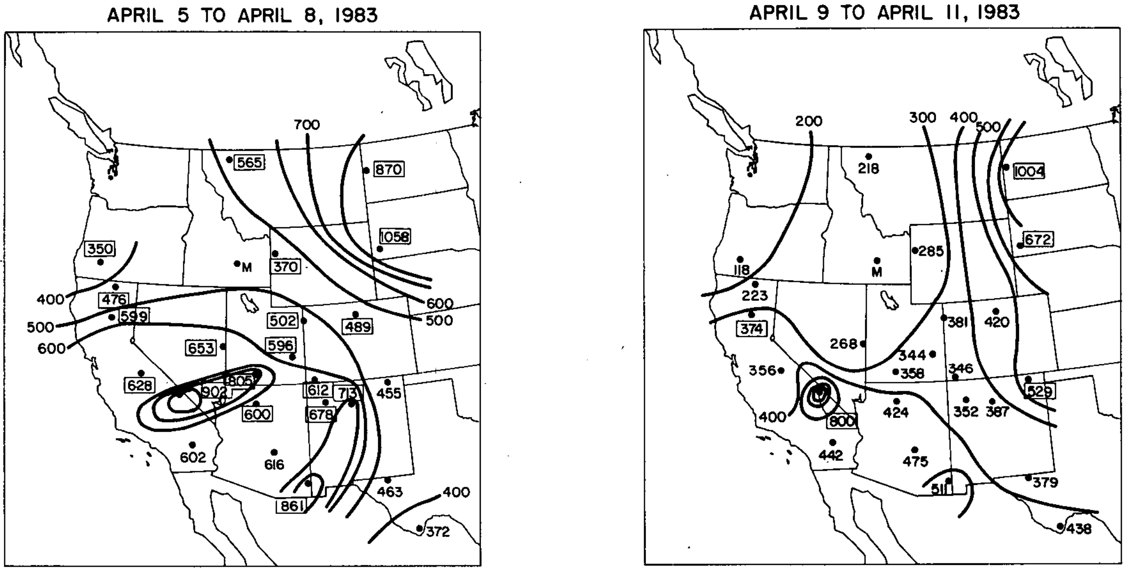

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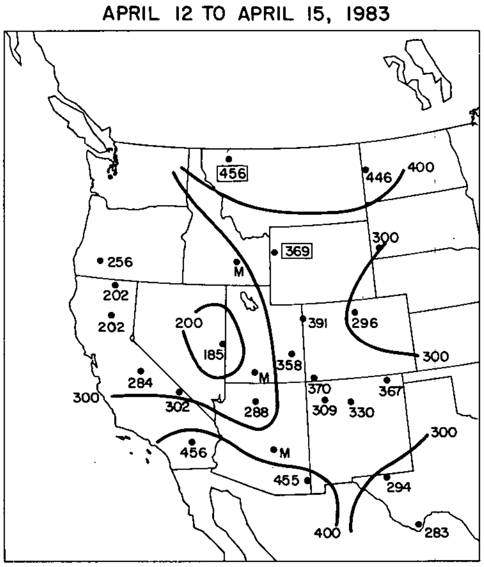

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FIG. 1. Fine particulate (diameters < 2.5 μm) sulfur concentrations in ng m^{-3} for the period: (a) 1200 LST 29 March to 1200 LST 1 April 1983, (b) 0000 LST 2 April to 0000 LST 5 April, (c) 1200 LST 5 April to 1200 LST 8 April, (d) 0000 LST 9 April to 0000 LST 12 April, and (e) 1200 LST 12 April to 1200 LST 15 April. Concentrations less than one standard deviation below the site's geometric mean are circled. Those greater than one standard deviation above the mean are enclosed by a square. Missing observations are indicated by an "M."

format from the National Climatic Data Center. Backward trajectories of 5 days duration from all the NPS sites were calculated every 6 h during the period of interest.

The ARL-ATAD model is a Lagrangian parcel model that contains a single, variable-depth transport layer. Trajectory segments are computed every 3 h using time- and distance-weighted averages of the nearest rawinsonde observations. For backward trajectories, no estimate of dispersion is made, so the trajectories are used for qualitative assessment of air-mass source regions. Raynor et al. (1983) have demonstrated the utility of this model in locating source regions of material transported over long distances. Because of the qualitative nature of the study, errors inherent in the trajectory calculations, such as those described by Sheih (1983), Reisinger and Mueller (1983), and Kuo et al. (1985), will not be considered important. However, the trajectories should be thought of as only indicating the general geographic area for the origin of an air mass.

3. The April 1983 case

Figure 1 contains plots of the fine sulfur concentrations observed at the western NPS sites for the period 29 March to 15 April 1983. Figure 1a presents concentrations before the onset of the episode. At several sites in and west of the Great Basin, concentrations are less than one standard deviation below the site's two-year geometric mean, indicating very clean air. Concentrations elsewhere were at or slightly below average. Synoptically, a cold front trailing from a surface low in Alberta, Canada, passed through the northern part of the network during this period, accompanied by westerly flow off the Pacific Ocean. South of this front, winds during this period were light and from a southerly or westerly direction.

Figure 1b shows the beginning of the episode in the northern part of the network. Sulfur concentrations exceeded one standard deviation above the mean at sites north of northern Colorado and east of Oregon. During this period, a strong (984 mb) surface low pressure center was traversing northeastward through the central United States, while an east–west elongated surface high pressure area was in south-central Canada. Such a synoptic pattern has been shown to be accompanied by high sulfur concentrations in the northern Plains (Henmi and Bresch, 1985). Trajectories for the period constructed backward from the receptor sites are shown in Fig. 2a. Over the Plains, trajectories were arriving from the east, north of the Midwest cyclone. At Craters of the Moon National Park, Idaho, westerly trajectories on 2 April switched to easterly on 3 April. If local sources are assumed negligible, then southwestward advection of fine particulate sulfur is implied by the sulfur concentration pattern and trajectory analysis. Over other portions of the network, trajectories were arriving from the west or north and were accompanied by average fine sulfur concentrations.

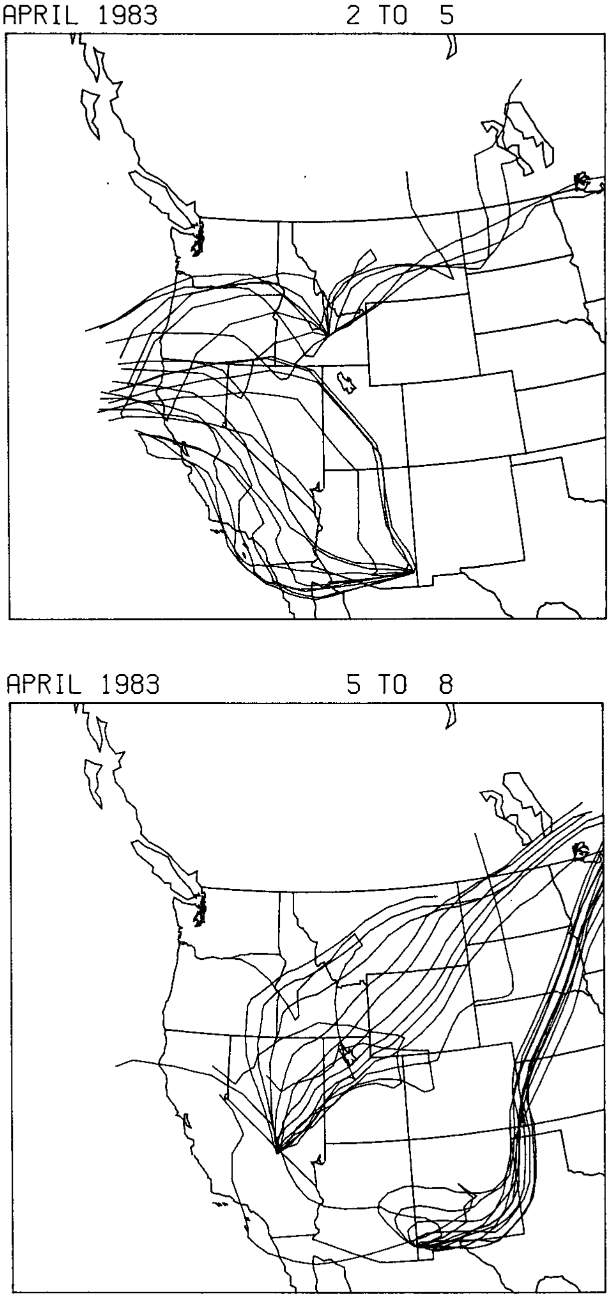

FIG. 2. Backward trajectories calculated by the ARL-ATAD model for (a) 2 to 5 April 1983 (corresponding to Fig. 1b) from Craters of the Moon National Park, Idaho, and Chiricahua National Monument, Arizona, and (b) 5 to 8 April 1983 (corresponding to Fig. 1c) from Death Valley National Monument, California, and Chiricahua National Monument, Arizona. Trajectories are started every 6 h and consist of 3 h segments, which are followed backward in time for 5 days.

During the period between 5 April and 8 April, fine sulfur concentrations were exceedingly high over al-

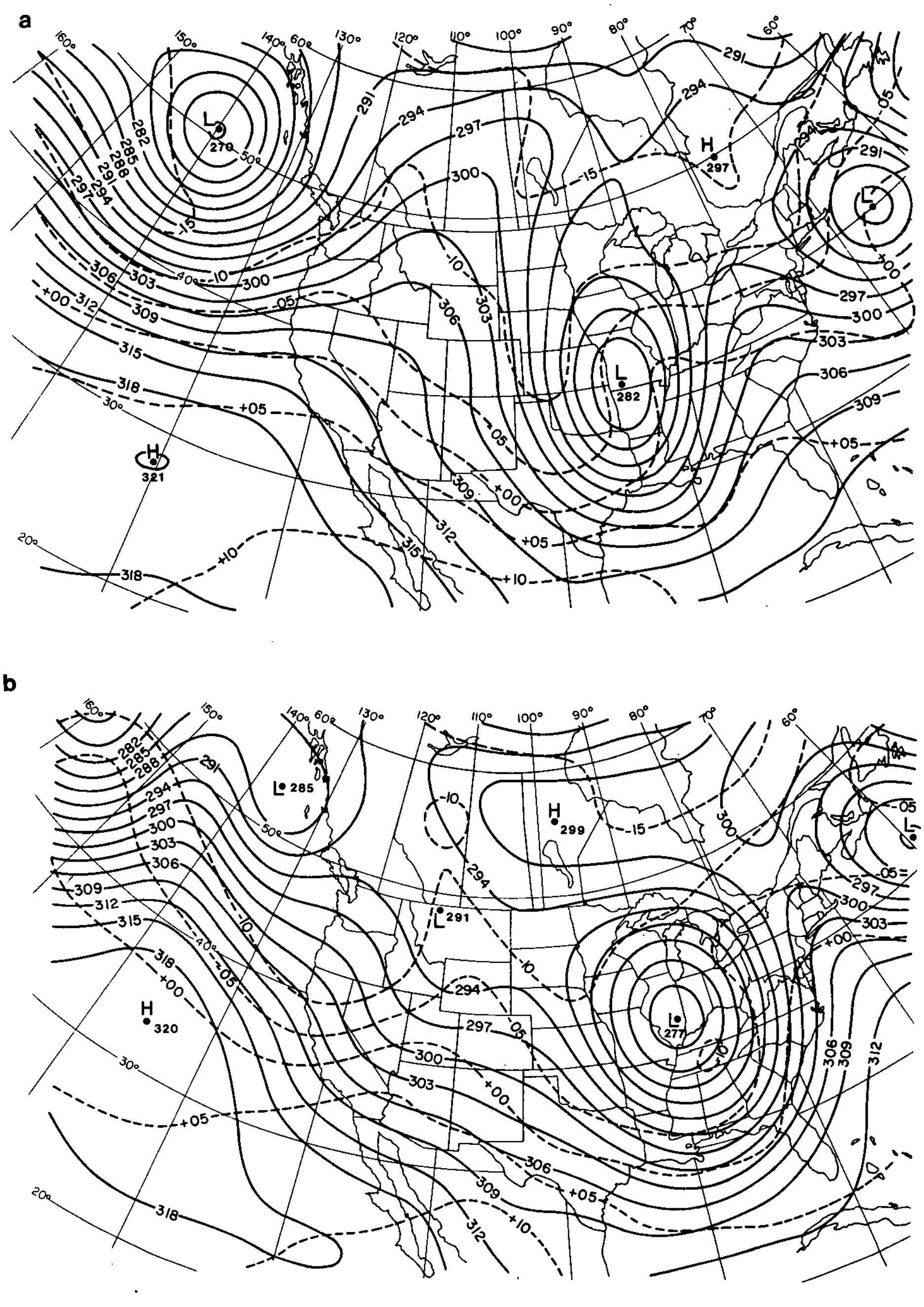

FIG. 3. The National Meteorological Center's analysis of 700 mb geopotential heights (dam, solid) and temperature (°C, dashed) for (a) 0000 UTC 2 April 1983, (b) 0000 UTC 3 April 1983, (c) 1200 UTC 4 April 1983, and (d) 0000 UTC 9 April 1983.

most all of the western United States (Fig. 1c). Figure 2b shows trajectories for this period. Air parcels at Death Valley National Monument in California were arriving from the northern Plains, and sites as far south as the Mexican border had trajectories arriving from the Midwest. At many of the sites, concentrations of fine particulate lead as well as soot increased substantially during this period; however, only at two sites, Bryce Canyon, Utah, and Grand Canyon, Arizona, did concentrations of fine copper and fine lead increase significantly. These two elements can be used as tracers of copper smelter emissions. Trajectories for these two sites (not shown) passed over the copper smelter near Salt Lake City.

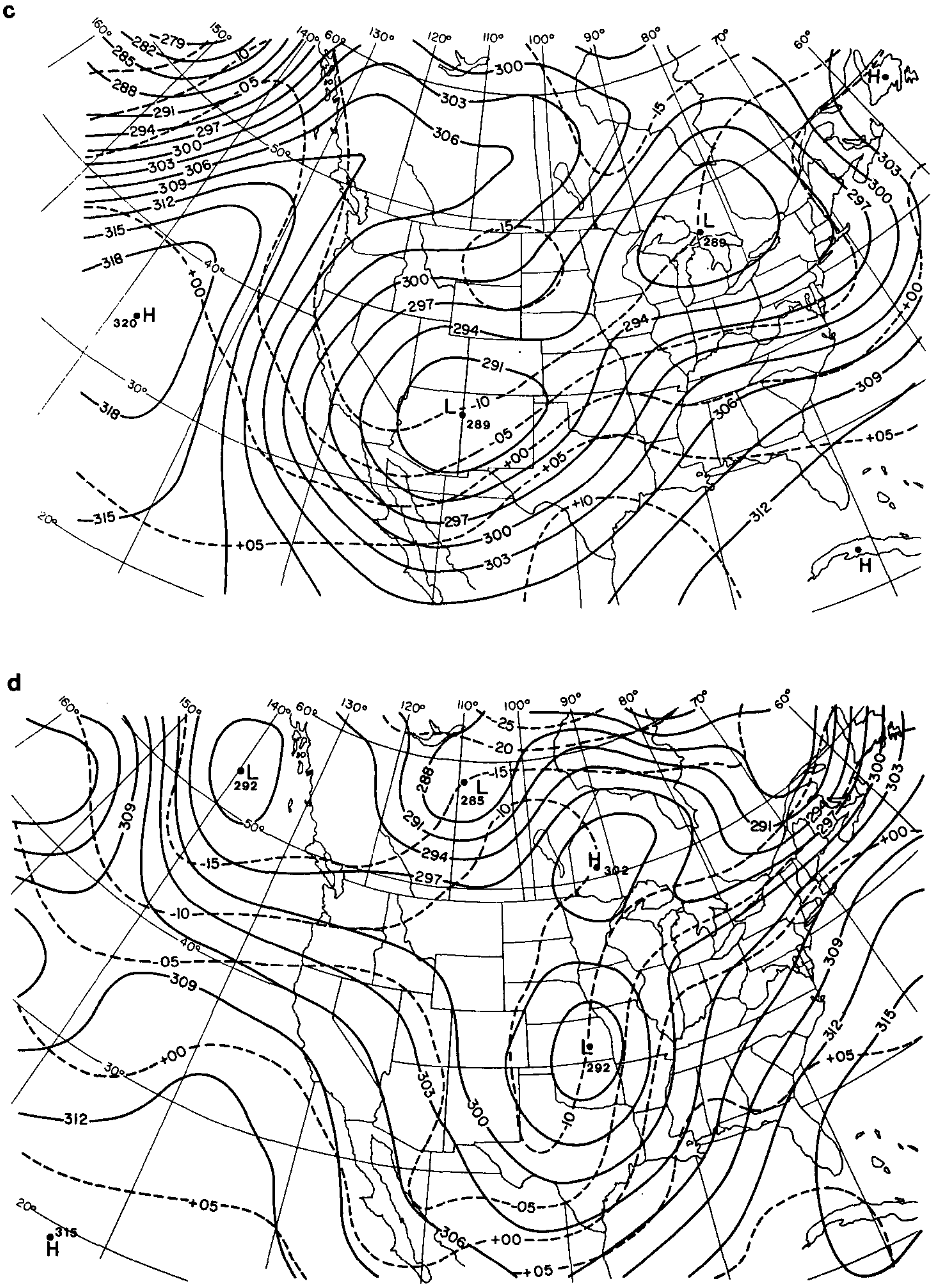

FIG. 3. (*Continued*)

The trajectories shown in Fig. 2b indicate that air parcels would have had to have crossed the Continental Divide. At Death Valley, trajectories from North Dakota arrived during the daylight hours on 6 April. The transport layer depth (TLD) as calculated by the ARL-ATAD model (see Heffter, 1980, for details) was near the model's maximum allowable value, between 2800 and 3000 m above the terrain, until the trajectories reached North Dakota, when the TLD dropped to approximately 1900 m. This deep transport layer may be attributed to unstable lapse rates associated with a cold upper low, as discussed below. Also, most of the trajectories crossed the Continental Divide in Wyoming, where the Divide is at its lowest elevation (less than 2100 m MSL).

Trajectories arriving at Death Valley on 6 April resided over the Dakotas on 3 April. Fine particulate sulfur concentrations over the Dakotas at this time were approximately 1000 ng m^{-3} (Fig. 1b). Trajectories from other sites in the West (not shown) such as Crater Lake, Oregon; Lehman Caves, Nevada; and Bryce Canyon, Utah; also arrived from Wyoming and the Dakotas during this period. Figure 1d shows the concentrations for the subsequent 3-day period. Concentrations began to drop from west to east as a Pacific cold front crossed the region. Apparently, the polluted air mass moved eastward, as indicated by the increase in sulfur concentration over the previous period at Capulin Mountain in northeast New Mexico.

The high concentration episode accompanied a major change in the tropospheric circulation. Figure 3 shows the 700 mb geopotential height and temperature fields for the period of interest. On 2 April (Fig. 3a) troughs were located over the central United States and near 50°N, 140°W with a ridge over the intermountain region. On 3 April, (Fig. 3b) a short wave from the Pacific trough began to break down the ridge. In response, the central United States trough began to retrograde so that by 4 April (Fig. 3c) a strong cutoff low was located over the southwestern United States and persisted for several days. Note the northeast flow across the Dakotas, easterly over Wyoming and Montana and continuing southwestward toward the California coast. The cold air aloft led to lapse rates over the Dakotas of 6.7°K km^{-1}. By 9 April, the low center had begun to move eastward (Fig. 3d).

The combination of easterly upslope flow, unstable lapse rates, and sufficient moisture led to widespread light snow over most of the higher elevation sites in Wyoming, Colorado, and New Mexico from 3 to 6 April. Even though this snow was falling through sulfur-laden air, National Atmospheric Deposition Program (NADP) wet deposition measurements of pH and sulfate in the melted precipitation over the region were not significantly different from their average values. This observation agrees with some recent observations showing that aerosols smaller than 10 μm are inefficiently scavenged by inertial impaction of falling snow crystals and that the acidity of snow crystals is not linearly related with the particulate loading of the air (Borys, personal communication, 1986).

From these analyses, the episode can be summarized as follows: The strong circulation around a Midwest cyclone on 2 April pumped relatively polluted air from the midwestern United States over the Great Lakes region and into the northern Plains. Concentrations at Shenandoah National Park, Virginia, which might be considered representative of concentrations in the Midwest, were approximately 1300 ng m^{-3} until 5 April. On 3 and 4 April the retrograding upper wave pattern drew in this polluted air from the upper Midwest toward the Pacific Coast. The polluted air lingered in the West until the upper low moved eastward on 9 April.

4. Summary and conclusions

Observations of extremely high fine particulate sulfur concentrations during early April 1983 in the western United States were linked to a strong cyclone over the midwestern United States. The strong winds around this cyclone circulated midwestern air (which typically has high sulfur concentrations by western standards) as far west as the Pacific Coast. A retrograding upper-wave pattern was conducive for this polluted air to move southwestward. Both a long-range trajectory analysis and a subjective evaluation of synoptic conditions confirm this hypothesis.

Although westward transport of polluted air from the Midwest to the Pacific Coast is not a common occurrence (this was the only clear-cut case in a two-year measurement period), emissions in the midwestern United States apparently can impact on the pristine areas of the West. Another similar episode occurred in April 1984, but missing particle data prevents a thorough analysis. Such transport occurs only during periods of strong tropospheric circulation in the spring and perhaps the fall. This easterly upslope flow is often accompanied by precipitation, but in this case wet deposition measurements were not significantly different from average, perhaps because of the poor scavenging properties of snow. Further studies are necessary to examine the impact of various source regions upon the deposition and pollutant concentrations in the West.

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