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THE INFLUENCE OF CUT-OFF LOWS ON SULFATE BURDENS OVER THE NORTH ATLANTIC DURING APRIL, 1987

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1. INTRODUCTION

Sulfate aerosol is of great interest because of its role a scattering solar radiation and modifying microphysical properties of clouds, with possible resultant influence on the earth's radiation balance and climate (Hansen et al., 2000; Haywood et al., 1999; Kiehl et al., 2000). This material derives mainly from anthropogenic emissions of sulfur dioxide (SO_2) followed by oxidation in the atmosphere to form sulfate aerosol, which in turn is removed mainly in precipitation. At any given time the distributions of SO_2 and sulfate are dependent on the synoptic-scale meteorological fields over the past several days. In view of the temporal variation in loadings that results from the variability in the controlling synoptic-scale meteorology, it is virtually mandatory for the purpose of performing short-term comparisons with observations to drive the models with observation-derived synoptic meteorological data.

We have previously described a model in Benkovitz et al. (1994), hereinafter B94, and Benkovitz and Schwartz (1997), hereinafter B97, that represents the distribution of SO_2 and sulfate for a domain covering North America, the North Atlantic and Europe and have presented snapshots of the mixing ratio and column burden showing the large variation in the distributions of these quantities at different times as a consequence of day-to-day differences in the governing synoptic-scale meteorology.

Here we present results of a 32-day simulation in March-April 1987, focusing on certain events for which we present a description of the governing synoptic-scale meteorology and its influence on the evolution of the chemical fields. We focus in particular on the role of cut-off low pressure systems and relate the distribution of SO_2 and sulfate to the responsible distribution of sources and the governing transport and transformation processes.

2. TRANSPORT AND TRANSFORMATION MODEL

The model used in this study, the Global Chemistry Model driven by Observation-derived meteorological data (GChM-O), is a three-dimensional Eulerian transport and transformation model for sulfur species. The physical and chemical mechanisms used in the model have been previously described in B94, and B97 and are briefly described here. The model represents emissions of anthropogenic SO_2 and sulfate and of biogenic sulfur species, horizontal and vertical transport, gas-phase oxidation of SO_2 and dimethylsulfide (DMS), aqueous-phase oxidation of SO_2 by hydrogen peroxide (H_2O_2) and ozone (O_3) in precipitating liquid water clouds, and wet and dry deposition of SO_2, sulfate, and methanesulfonic acid (MSA). The SO_2 and sulfate variables are tagged according to source (North America, Europe, and biogenic). The model is driven by the 6-h uninitialized analysis from the European Centre for Medium-Range Weather Forecasts (ECMWF, 1988); the horizontal coordinates are latitude and longitude with 1.125° resolution and the vertical coordinate is the ECMWF terrain-following η (eta) coordinate.

Anthropogenic emissions of sulfur species in the model were taken to be representative of the March and April time frame circa 1985. Marine biogenic emissions of DMS (Bates et al., 1992) were gridded based on a global monthly gridded pigment concentrations derived from Coastal Zone Color Scanner (CZCS) images; gridded terrestrial biogenic emissions were calculated using the methodology of Lamb (Bates et al., 1992) and treated entirely as DMS. Biogenic emissions in the model domain are approximately 5% of the anthropogenic emissions.

Time- and location-dependent dry deposition of SO_2 and accumulation mode particulate sulfate are included in the model; deposition velocities were calculated from surface resistance, atmospheric stability, and surface wind speeds (Wesely, 1989). Dry deposition is an important removal mechanism for SO_2 but is minimal for sulfate, with an effective first-order removal rate of 5% per day at most. Gas-phase oxidation of SO_2 and DMS is represented as pseudo first-order reactions using climatological diurnal-average OH mixing ratios from Spivakovsky et al. (1990). Oxidation of SO_2 by hydrogen peroxide (H_2O_2) was limited by the lesser of either the SO_2 or the H_2O_2 mixing ratio. H_2O_2 was advected in the same way as the sulfur species, depleted by reaction with SO_2, and regenerated in the gas phase at a fixed rate (0.021 ppb h^{-1}) until a maximum mixing ratio (1.4 ppb) was reached. Aqueous-phase oxidation of SO_2 by ozone (O_3) was represented by a pseudo-first-order reaction of gaseous SO_2 with a rate constant that takes into account an assumed O_3 mixing ratio of 37.5 ppb, O_3 solubility, the solubility and dissociation of gaseous SO_2, and the cloud liquid water content; cloudwater pH was taken as 4.5.

3. THE INFLUENCE OF METEOROLOGY ON SULFUR BURDENS

During the spring season rapid fluctuations occur between different types of synoptic regimes such as high amplitude meridional meanders, cut-off lows, and zonal flow. The connection between evolving meteorological flow features and the SO_2 and sulfate distributions is examined using meteorological analyses from two

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pressure levels in the troposphere, 925 hPa and 500 hPa. The gridded data used in the synoptic analyses were generated from the NCAR Reanalysis Project (Kalnay et al., 1996). As noted above, the SO_2 and sulfate simulations were driven by the ECMWF uninitialized analyses. The same underlying synoptic data are used in both analyses, although there are inevitably differences between the meteorological fields in the two analyses and subtle differences in the hemispheric height (wind) fields resulting from differences in the objective analysis schemes. The similarity of the meteorological fields suggests that these differences do not significantly impact the interpretation of the governing transport processes.

Horizontal height gradients on a constant pressure surface are used to depict the structure of the large-scale circulations that modulates the transport of trace species. Representative average heights above sea level are 5420 m for 500 hPa and 780 m for 925 hPa. Positive and negative height anomalies on the constant pressure surfaces correspond to high and low pressure systems that give rise to clockwise (anticyclonic) and counterclockwise (cyclonic) circulations respectively. Vertical material transport in cyclones is characterized by large-scale upward motion and abundant precipitation. Conversely, vertical material transport in anticyclones is characterized by downward motion, absence of precipitation, and strong low-level inversions. When cyclones decay, the surface and upper air circulations become vertically aligned.

Occasionally a system develops that is completely separated from the jet stream, manifested as concentric height contours surrounding a height minimum on the 500 hPa analysis. These systems, known as cut-off lows, can control the transport, removal, and accumulation of material over a region for periods of a week or longer as the circulations around them prevent ventilation by air masses from other locations.

High sulfate column burdens near the surface have previously been associated with anticyclones and resultant stagnation episodes (Husar and Wilson, 1993; Husar et al., 1997; Lyons, 1980). The present model results suggest that large sulfate column burdens may also occur in association with the decaying stages of certain cyclones, depending on their location relative to emission sources. Here we examine two examples of the different effects of cut-off lows on sulfur burdens over the North Atlantic Ocean.

3.1 1 April 1987

The 925 hPa and 500 hPa meteorological analyses for 1 April 0000UT, shown in Figure 1, reveal a well-defined cut-off low located at 50°W and 30°N vertically aligned at 500 hPa with a weak low pressure center at 925 hPa; the main axis of the jet stream roughly parallels the 5400 m height contour. The SO_2 column burden (vertical integral of the concentration) is small, as seen in Figure 2, because the cut-off low is far removed from local sources and the transit time allows for conversion to sulfate. Precipitation has removed sulfate in this system, although clearly the precipitation rate is not as large as in a warm-conveyor belt. This maritime system is spinning down and does not have strong dynamic forcing; thus the cut-off low acts as a sink for sulfur.

A second low pressure system over Italy is indicated by the height minimum in the synoptic analysis, with nearly concentric 500 hPa height contours and the corresponding low values. The synoptic analysis at 925 hPa shows a weak minimum in the height field vertically aligned with the height minimum at 500 hPa. Like the cut off low in the central Atlantic this disturbance is separate from the main flow at 500 hPa. However, in contrast this cut-off low is located over a continental

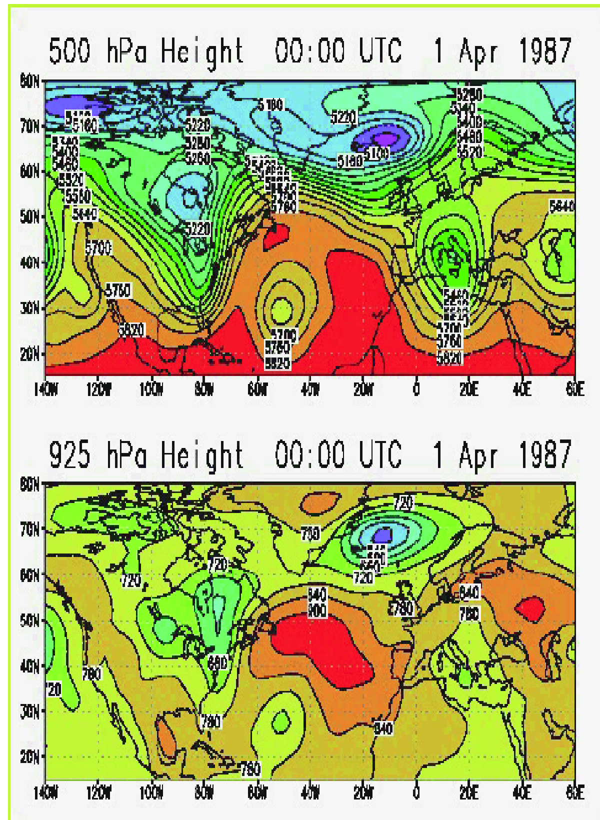


Figure 1. Height in meters of the 925 hPa and 500 hPa isobaric surfaces for April 1,1987 at 00UT, derived from synoptic analysis of the NCAR Reanalysis Project (Kalnay et al., 1996).

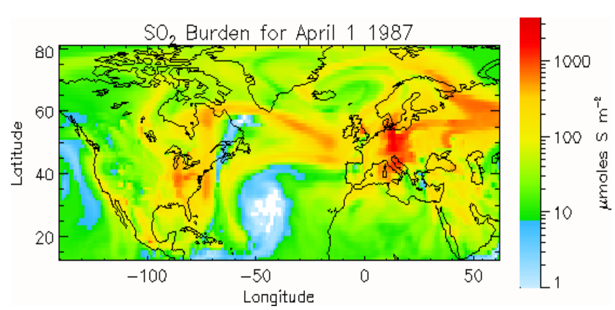


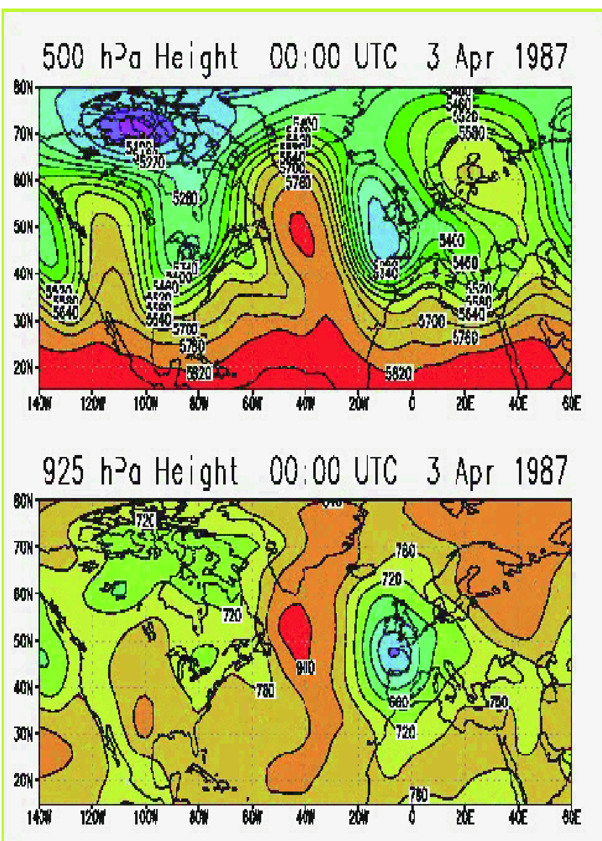
Figure 2. SO_2 column burden for April 1, 1987 at 00UT. Column burden is evaluated as the vertical integral of the concentration.

region with abundant sources of SO_2. The showery precipitation is less intense than in the previous case due to less moisture availability, so SO_2 from the local sources accumulates and is oxidized to sulfate.

In summary, the synoptic analysis for 1 April shows two physically similar cut-off lows which have markedly different impacts on the atmospheric sulfur burden due to their geographic location in relation to sources of moisture and sulfur emissions.

3.2 3 to 5 April 1987

We now focus on two cut-off lows observed on 3-5 April: one along the eastern margin of the Atlantic Ocean, impacting western Europe, and the other along the eastern seaboard of the United States, as seen in Figure 3. The 500 hPa surface on 3 April shows the development of a high-amplitude trough off western Europe with an associated strong surface storm seen in the 925 hPa panel. Subsequent panels show the evolution of this high amplitude trough into an intense cut-off low. During this period the air mass from the European continent is drawn into the developing cut-off low, as seen in Figure 4 which shows the development of the sulfate burdens over the region. The dry air is eventually moistened as it wraps around the system; by April 5 the sulfate column burden is up to several hundred μmol m^{-2} south of the low. These high values persist in the region for more than 72 hours. In these two cases the location of sulfur emission sources relative to the cut-off



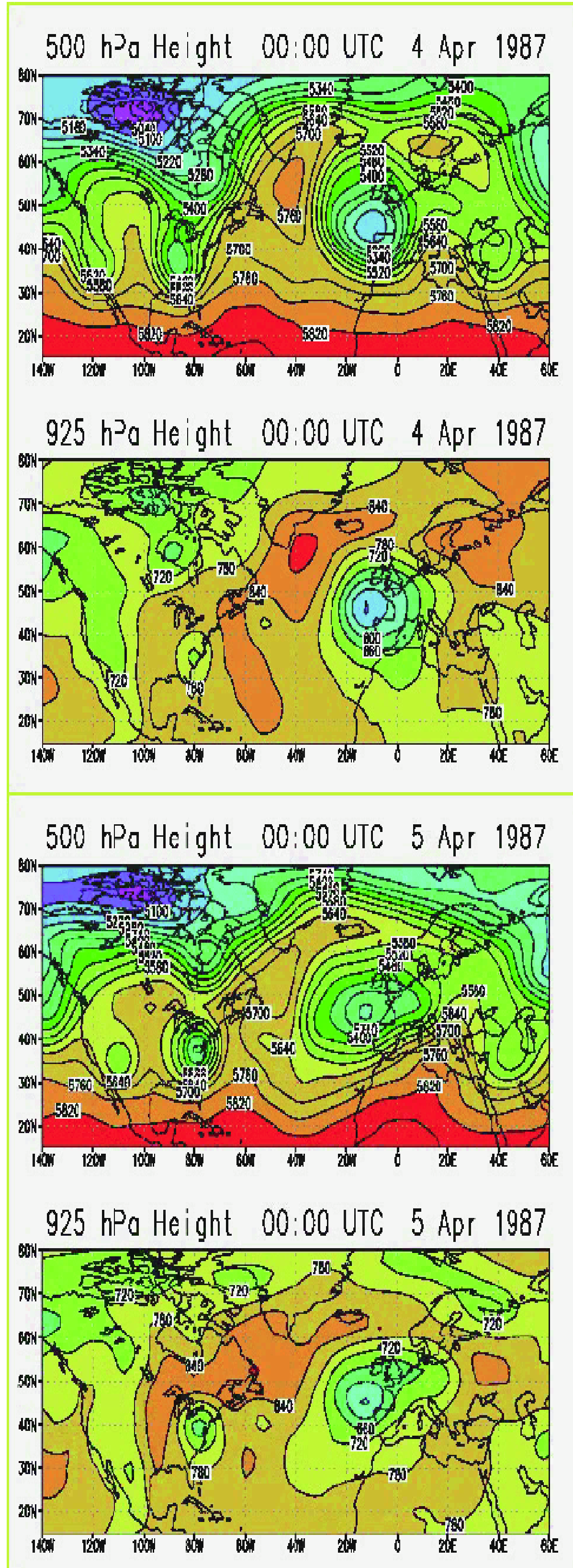


Figure 3. Synoptic analysis for April 3 to 5, 1987 at 00UT, as per Figure 1.

low result in quite different sulfate mixing ratios. In the low along the US seaboard precipitation quickly removes the sulfate produced, whereas in the low over western Europe the sulfate produced remains in the atmosphere for a relatively long time.

Without elaborating on the development phases of the two cut-off lows we note that on 4 April the size of these two low-pressure systems is comparable. However, in their more mature phases seen on the the 5 April analysis there is a distinct difference in size, with the low along the US seaboard being considerably smaller. Many of the dynamic features of the two lows are similar, including the circulations, yet the sulfate mixing ratios are markedly different. There are regions of the western European low where the sulfate mixing ratio exceeds 1 nmol/mol air (1 ppb), whereas the mixing ratio in the US low is an order of magnitude lower.

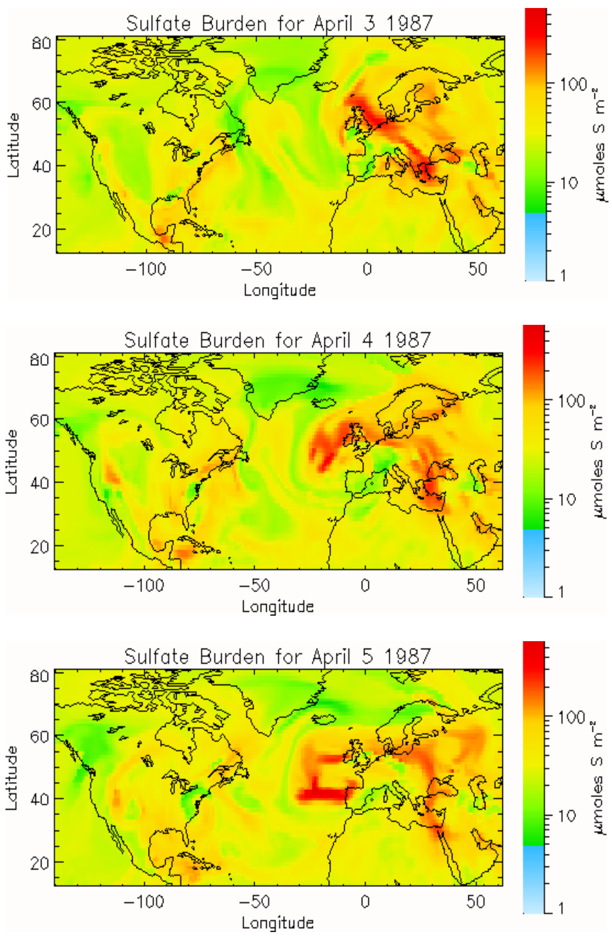


Figure 4. Sulfate column burden for April 3 to 5, 1987 at 00UT.

The low along the Western European coast exhibits concentric circulation, with little slantwise transport into or out of the core of the disturbance. This is an indication that the disturbance is in the final phase of its life cycle, an observation consistent with the lack of precipitation production and its vertical alignment on all isentropic surfaces. Such a configuration suggests that the same air mass is being continuously re-circulated over western European sulfur sources, the re-visitation rate being on the scale of approximately one day. This type of structure is consistent with the observed increase of the sulfate column burden from 3 April to 5 April, the vertical diffusion from surface sources evidently occurring in the form of SO_2 transport and subsequent conversion to sulfate. The variable sulfate loading during this episode has been used by Harshvardhan et al. (2001) to identify the influence of sulfate on cloud microphysical and radiative properties.

The difference in precipitation production between the cut-off lows along the US eastern seaboard and over western Europe is related to the geographical position of the low pressure systems relative to sources of moisture. Dynamics dictate that the slantwise ascent in these systems, before they reach the cut-off stage, takes place on the eastern side of the disturbance that marks the low. The likelihood of the parcels undergoing slantwise ascent eventually producing precipitation depends upon the availability of surface moisture at the start of the ascent. The low over the eastern seaboard of the US is lifting moisture-laden parcels originating over the warm Gulf of Mexico and the Atlantic Ocean, generating in precipitation which removes sulfate; the low over western Europe is lifting dry parcels originating from cold land surfaces over continental Europe, giving little opportunity for precipitation and sulfate removal.

4. SUMMARY AND CONCLUSIONS

We have presented examples from a modeling study of the development of sulfur burdens over North America, the North Atlantic Ocean and Europe during April, 1987 using observation-derived meteorological data to represent the actual conditions for this period, focusing on the influence of cut-off lows on SO_2 and sulfate column burdens over the North Atlantic Ocean. The analysis demonstrates that these systems can serve either as sources or sinks of sulfate, and that the major factor governing their resulting effect is the position during its formative stages relative to a) sources of moisture, and b) sulfur emissions, which regulates the availability of sulfur, cloud liquid water for sulfur oxidation, and the amount of precipitation for sulfate removal produced in the later stages of the life cycle.

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