Turnover Times and Mean Heights of Modeled Sulfate and SO_2 over the North Atlantic and Adjacent Continental Regions

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Abstract

Shortwave radiative forcing of climate by direct light scattering by sulfate and other aerosols is considered important in the context of anthropogenic climate change. Representation of this forcing in climate models requires a chemical model for the loading and distribution of this aerosol whose accuracy must be evaluated by comparison with observations. We have previously described a model for sulfate aerosol driven by observation-derived synoptic-scale meteorological data suitable for such comparisons (Benkovitz et al. 1994) and presented such comparisons for four seasonal months in 1986-1987 (Benkovitz and Schwartz 1997). Here we examine turnover times, mean heights, and other properties of SO_2 and sulfate from anthropogenic sources in North America and Europe and of sulfate and methanesulfonic acid (MSA) from biogenic sources. We have generated maps showing the geographical distribution and mean height of sulfate from the several sources. In general, North American sulfate dominates over the mid-North Atlantic, but occasionally there are substantial incursions of European sulfate. Primary sulfate is generally at lower altitude (mean height mainly ~1 km to 2 km) than secondary sulfate (~3 km to 5 km). Sulfate turnover times are typically about 5 days. Decorrelation distances of sulfate column burden are less than 1000 km, and decorrelation times less than 24 h, even in the mid North Atlantic well removed from sources.

Introduction

Tropospheric aerosols are important agents of radiative forcing of climate and must be accurately represented in climate models. This requires accurate models for loading and distribution of tropospheric aerosols. Sulfate aerosols are implicated as major contributors to aerosol climate forcing over the industrial period. We have previously described a subhemispheric scale transport and transformation model for tropospheric sulfate aerosol driven by observationally derived synoptic meteorological data (Benkovitz et al. 1994). Results have been presented for runs covering four seasonal months in 1986-1987, including comparisons with surface measurements of sulfate and SO_2 mixing ratios from monitoring networks (Benkovitz and Schwartz 1997). Concentrations or column burdens of anthropogenic atmospheric aerosols, such as sulfates, are highly variable spatially and temporally because of the short residence times of the aerosol particles relative to the time scale of variability of synoptic-scale meteorology that is responsible for their transport and deposition, and because of the highly nonuniform distribution of sources. This temporal and spatial variation is readily visualized in animations of column burden of sulfate, available on the web at *http://www.ecd.bnl.gov/sulfate_model.html*

This Study

Here we examine turnover times, mean heights, and other properties of SO_2 and sulfate from anthropogenic sources in North America and Europe and of sulfate and MSA from biogenic sources.

Key Findings

Following are key findings from the study:

- Turnover times of sulfate are typically about 5 days, consistent with values that have been employed in previous estimates of sulfate aerosol forcing (Figure 1). Turnover times of SO_2 and dimethylsulfide (DMS) are generally 1 to 2 days, but longer in winter when photochemistry and dry deposition are reduced.

- Both the column burden of sulfate and its mean height exhibit substantial spatial variation at a given time (Figure 2).

- In general, North American sulfate dominates over the mid-North Atlantic, but occasionally there are substantial incursions of European sulfate.

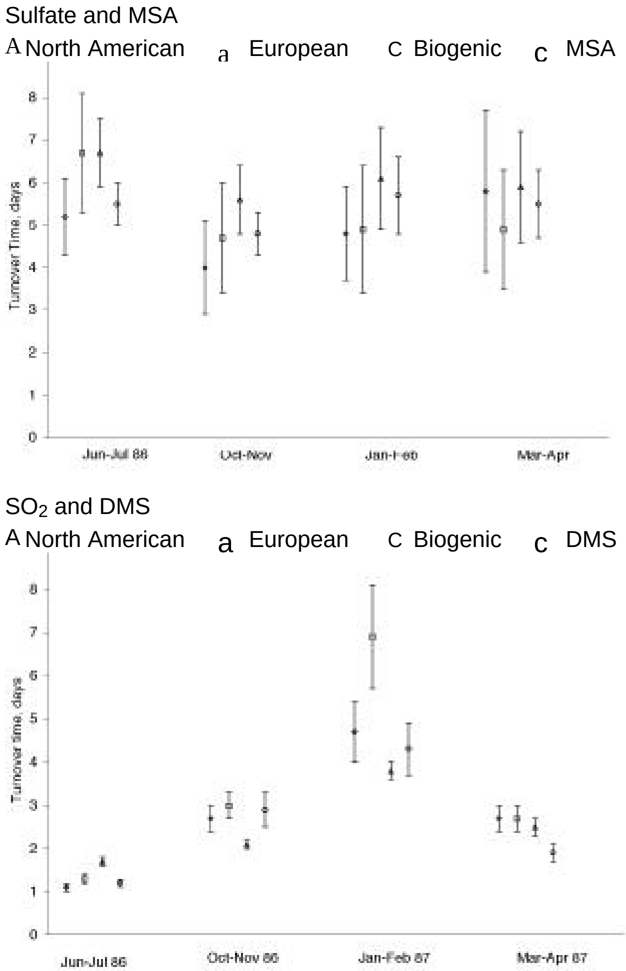

Figure 1. Turnover times of sulfur species evaluated from model runs for four seasonal months in 1986-87.

- Primary sulfate is generally at lower altitude (mean height mainly ~1 km, except summer, ~2 km) than secondary sulfate (~3 km; 4 km to 5 km summer) (Figure 3).

- Decorrelation distances (the distance for the spatial autocorrelation coefficient of column burden at a given time to drop to 1/*e*) are less than 1000 km and decorrelation times (the time for the temporal autocorrelation coefficient of column burden at a given location to drop to 1/*e*) are less than 24 h, even in the mid-North Atlantic well removed from anthropogenic sources.

These findings suggest that it is necessary to take short time and space variation into account in comparisons of measured and modeled aerosol loadings and in evaluation of forcing, especially the indirect forcing, which is highly nonlinear in aerosol loading.

References

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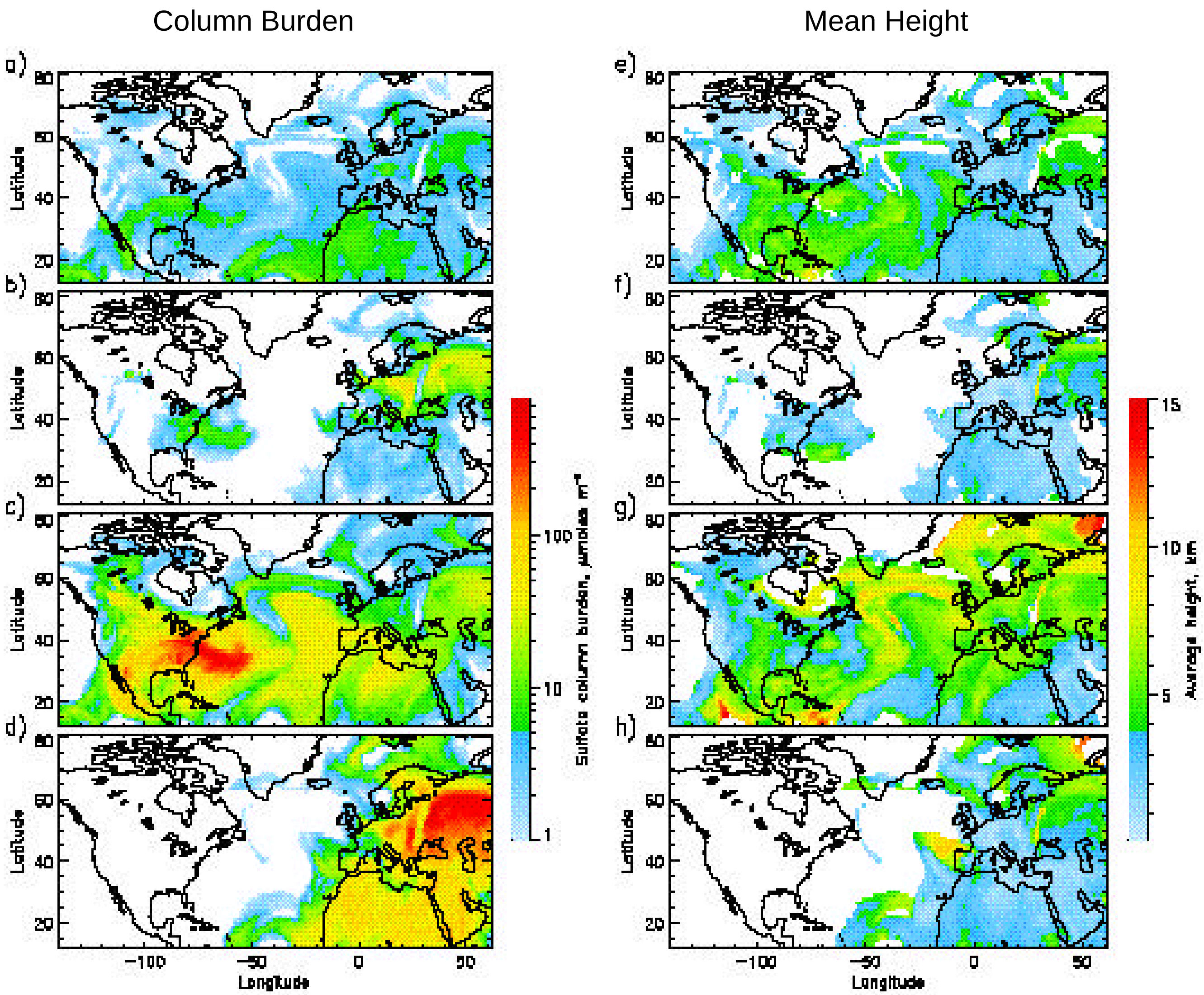

Figure 2. Column burdens and mean heights of sulfate in the model domain evaluated for July 9, 1986, at 0000 UTC. From top, biogenic sulfate, primary sulfate, secondary sulfate from North American emissions and secondary sulfate from European emissions. (For a color version of this figure, please see *http://www.arm.gov/docs/documents/technical/conf_9803/schwartz(2)-98.pdf*.)

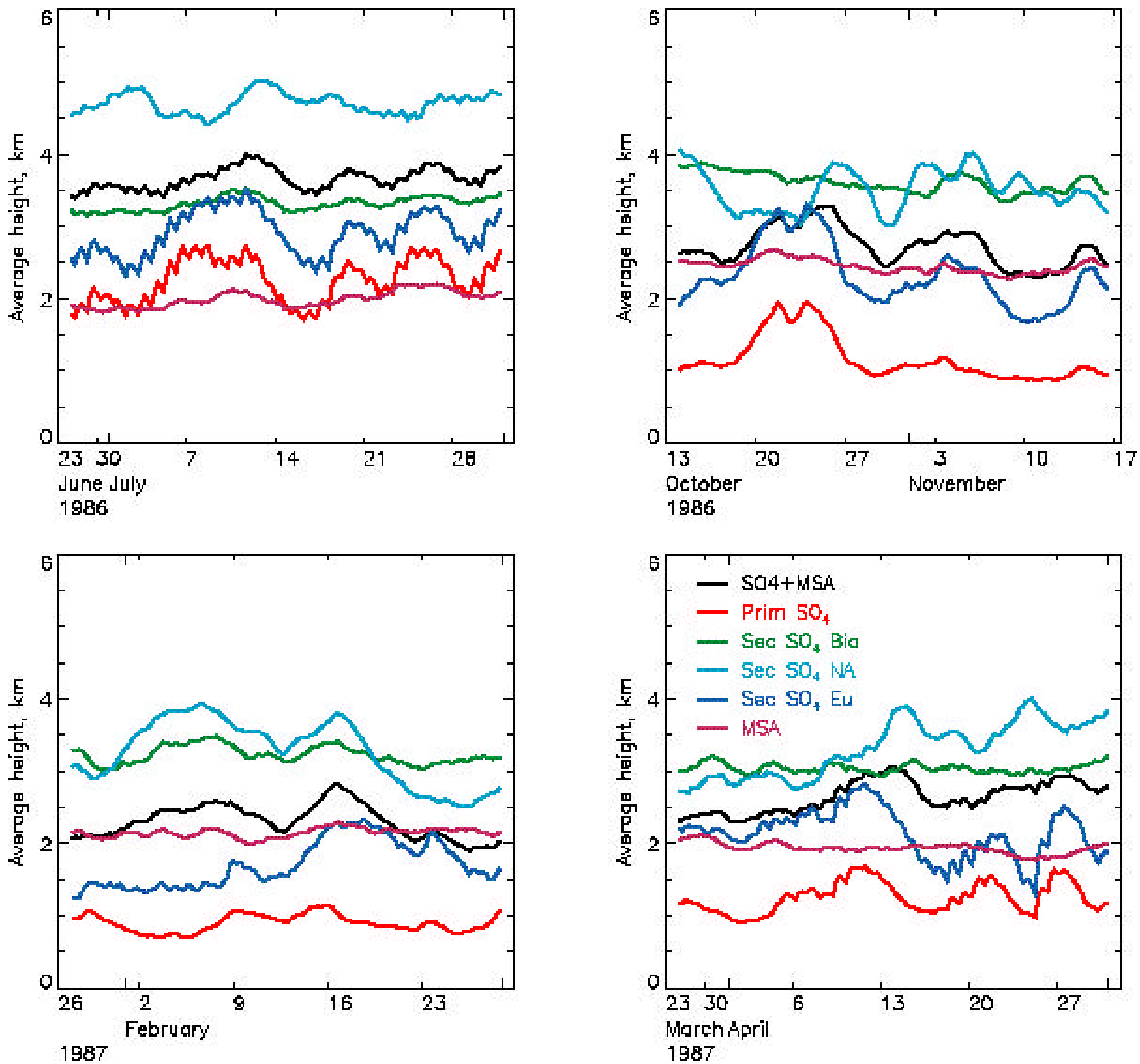

Figure 3. Mean height of the several sulfate species as a function of date for the four months of simulation. (For a color version of this figure, please see *http://www.arm.gov/docs/documents/technical/conf_9803/schwartz(2)-98.pdf*.)