Investigation of Trends in Aerosol Direct Radiative Effects over North America Using a Coupled Meteorology-Chemistry Model

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Abstract A comprehensive investigation of the processes regulating tropospheric aerosol distributions, their optical properties, and their radiative effects in conjunction with verification of their simulated radiative effects for past conditions relative to measurements is needed in order to build confidence in their estimates of the projected impacts on future climate. This study aims at addressing this issue through a systematic investigation of changes in anthropogenic emissions of SO_2 and NO_x over the past two decades in the United States, the consequent changes in anthropogenic aerosol loading in the North American troposphere, and subsequent impact on regional radiation budgets.

Keywords: Coupled meteorology-chemistry models, aerosol radiative effects

Introduction

A key uncertainty in quantification of aerosol radiative effects and their impact on climate change is the verification of the spatial and temporal variability in its magnitude and directionality and, consequently, its cumulative effect on the radiation balance of the earth-atmosphere system. While global numerical atmospheric models have recently been used to estimate the anthropogenic aerosol radiative effects for future changing emission scenarios, little effort has been devoted to verifying the fidelity of the simulated radiative effects relative to available observations. A key outstanding question in addressing these uncertainties is how well do current models represent the regional and temporal variability of aerosol radiative forcing for current and past conditions?

There is increasing evidence that the amount of solar radiation incident at the Earth's surface is not stable and has undergone significant decadal variations. The variability in this surface solar radiation (also referred to as surface shortwave radiation) plays a prominent role in the earth's climate system as it is the source

for energy exchanges between the atmosphere, and the land/biosphere/ocean components, and consequently can contribute to the modulation of the surface temperature, intensity of the hydrological cycle, and potentially the net ecosystem productivity (Wild, 2009). Analyses of long-term records of surface radiation measurements have suggested a decreasing trend during the 1960- 1980s time period, followed by period of leveling-out and to reversal (brightening) in the 1990s and thereafter. The documented long-term trends in surface solar radiation and their association with trends in emissions of aerosols and precursor species provide cases to test the representation of aerosol-radiation effects in current and evolving regional and global coupled chemistry/climate models.

Datasets and Model Overview

Title IV of the Clean Air Act Amendments (also known as the Acid Rain Program) has achieved substantial reductions in U.S. electric power industry emissions of SO_2 and NO_x since the 1990s. Analysis of ambient measurements of gaseous and particulate phase sulfur concentrations at CASTNET and IMPROVE sites show significant decreasing trends during the 1995-2010 period. Observations of both all-sky and clear-sky surface short wave radiation at many sites in the eastern U.S. show an increasing trend during this period.

The coupled WRF-CMAQ modeling system (Mathur et al., 2009; Wong et al., 2012) is exercised for selected summer time slices (June-July-August for the years 1990, 1996, 2000, 2006) to simulate the changes in tropospheric aerosol loading and associated radiative effects resulting from the changes in anthropogenic emissions over the past two decades. Model simulations were performed over two domains: an outer hemispheric scale domain covering the Northern hemisphere, set on a polar stereographic projection and discretized with a horizontal resolution of 108 km (cf. Mathur et al., 2012) and a nested 12 km resolution grid covering the Continental U.S. and portions of Canada and Mexico. Year specific emissions for the norther hemispheric domain were derived from the EDGARv4.2 global emission inventory. The regional 12 km resolution calculation employed emission estimates from recently developed inventory for 1990-2010 in which state-level anthropogenic emissions of SO_2, NO_x, CO, NMVOC, NH_3, PM_{10} and $PM_{2.5}$ for a total of 49 sectors were estimated based on several long-term databases containing information about activity and emission controls (Xing et al., 2013).

Discussion

Simulated changes in tropospheric aerosol levels for different regions of the northern hemisphere are illustrated in Figure 1 which shows summertime regional mean SO_4^{2-} concentrations across the eastern U.S., western Europe, and eastern

China. Systematic reductions in ambient SO_4^{2-} levels in the east U.S. and western Europe are noted for the 1990-2006 period and are attributed to SO_2 emission reductions in these regions. In contrast a significant increase in SO_4^{2-} levels across east China are noted post-2000 resulting from increase in industrial activity and energy demand and associated SO_2 emissions. These heterogeneities in changing tropospheric aerosol burden are likely to result in differences in regional radiation perturbation across these regions.

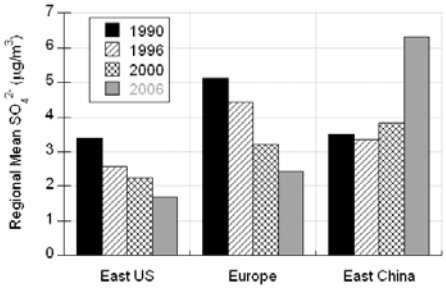

Figure 1: Simulated regional summertime mean SO_4^{2-} concentrations

The simulated reductions in ambient SO_4^{2-} over the eastern U.S. are consistent with trends in measured surface SO_4^{2-} concentrations at several sites in the region (Gan et al., 2013). Figure 2a presents spatial distributions of the modeled trends

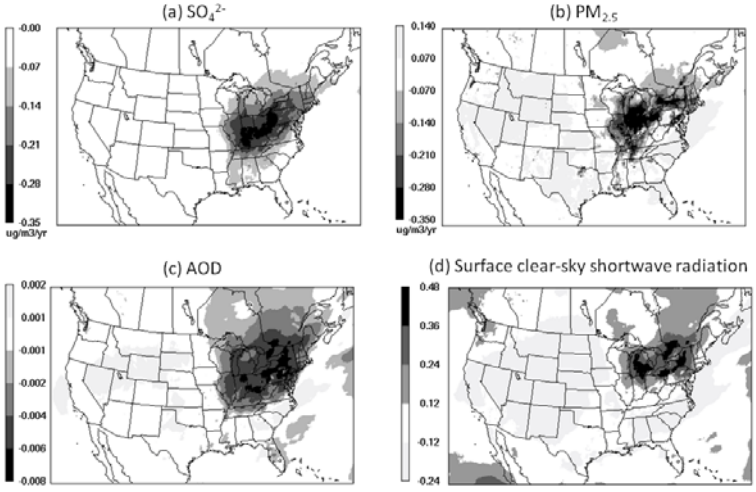

Figure 2: Spatial distribution of modeled trends during 1990-2006 in: (a) surface-level SO_4^{2-} ($\mu g/m^3/yr$), (b) surface-level $PM_{2.5}$ ($\mu g/m^3/yr$), (c) aerosol optical depth, and (d) surface clear-sky shortwave radiation($W/m^2/yr$). All trends based on summer (June-July-August) values.

in surface-level SO_4^{2-} inferred from a linear regression of simulated summer (June-July-August) average values for the four simulation years, 1990, 1995,

2000, 2006. Largest reductions in SO_4^{2-} (>0.3 μg/m^3/yr) are estimated downwind of the Ohio River valley where many electric generation units are based. The model estimated values in Figure 2a are in the range of those inferred from observations in the eastern U.S. (-0.093 – -0.14 μg/m^3/yr) as detailed in Gan et al. (2013). Regional reductions in ambient $PM_{2.5}$ are also simulated across the east U.S for the period 1990-2006 (Fig. 2b). The corresponding changes in aerosol optical depth is illustrated in Fig. 2c which shows a systematic decreasing trend in tropospheric aerosol burden across the eastern U.S. Associated with the reductions in aerosol burden in the region is a corresponding increase in simulated clear-sky surface shortwave radiation. Based on analysis of measurements at 3 eastern U.S. SURFRAD sites and the SGP site, we estimate an average increasing trend of 0.37 W/m^2/yr in clear-sky shortwave radiation during 1995-2010 (Gan et al., 2013). The corresponding model estimated trend is comparable in magnitude to the observed values and thus provide an estimate of the changes associated with direct aerosol radiative forcing in the east U.S.

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Questions and Answers

Questioner Name: Nicolas Moussiopoulos

Q: How do your model results regarding the direct effects compare against model uncertainty?

A: Unfortunately very few concurrent measurements of atmospheric aerosol size and composition profiles, their optical properties, and radiation are available to conduct a comprehensive closure experiment. We have used measurements from recent field campaigns to verify the model aerosol optics calculations and establish its credibility. We have also compared simulated surface solar radiation with measured values, under both moderate and high pollution episodes (e.g., wildfires) and found that including direct effects of aerosols helps improve the predictions of surface solar radiation. The long-term simulations discussed here are in fact another approach to build confidence in model predictions through comparison of trends for past conditions.

Questioner Name: Gabriele Curci

Q: In the comparison of ΔAOD and corresponding ΔADRE it looks like positive ΔAOD have larger impact on ΔADRE than negative changes: any hints why this is?

A: The aerosol optical properties (and consequently AOD) and their radiative effects are non-linearly dependent on a number of factor including aerosol size, composition, water content, albedo. The change in aerosol direct radiative effects (ΔADRE) thus should not be expected to strictly vary linearly with ΔAOD.

Questioner Name: Yang Zhang

Q: In analyzing the trend in observed PM between 1990 and 2006, have the PM observations been adjusted for meteorological differences between a specific year and climatology?

A: In deriving the trends in observed PM, we did not perform any adjustment to account for year-to-year differences in meteorology. The model simulations for only the summer months for just four selected years in the 1990-2006 period presented here are preliminary, and are clearly not sufficient to infer a robust trend. We are currently completing model calculations for several 3-year long time slices during 1990-2010. We will then average both the model and the measurements for the appropriate periods to infer trends that can be cross-compared in a consistent manner.