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Effects of emissions change, climate change and long-range transport on regional modeling of future U.S. particulate matter pollution and speciation

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ABSTRACT

This study investigates the future U.S. $PM_{2.5}$ pollution under multiple emissions scenarios, climate states, and long-range transport (LRT) effects using the regional Community Multi-scale Air Quality (CMAQ) model integrated with a regional climate model. CMAQ with fixed chemical lateral boundary conditions (LBCs) successfully reproduces the present-day $PM_{2.5}$ pollution and its major species in rural and suburban areas, but has some discrepancies in urban areas such as the Los Angeles Basin, where detailed emissions and meteorology conditions cannot be resolved by the 30 km grid. Its performance is slightly worsened when using dynamic chemical LBCs from global chemical transport model (CTM) simulations, which provide cleaner conditions into the CMAQ lateral boundaries. Under future Intergovernmental Panel on Climate Change (IPCC) emission scenarios, CMAQ projects large $PM_{2.5}$ reductions (~40% for A1B and ~20% for A1Fi scenario) in the eastern United States, but slight to moderate increases (~5% for A1B and ~10% for A1Fi) in the western United States. The projected increases are particularly large (up to 30%) near the Mexico-U.S. border, suggesting that Mexico is a major source for future U.S. $PM_{2.5}$ pollution. The effect from climate change alone is estimated to increase $PM_{2.5}$ levels ubiquitously (~5% for both A1B and A1Fi) over the United States, except for a small decrease in the Houston, Texas area, where anthropogenic non-methane volatile organic compounds (NMVOCs) emissions dominate. This climate penalty, however, is substantially smaller than effects of emissions change, especially in the eastern United States. Future $PM_{2.5}$ pollution is affected substantially (up to −20%) by changes in SO_2 emissions and moderately (3–5%) by changes in NO_x and NH_3 emissions. The long-range transport (LRT) effects, which are estimated by comparing CMAQ simulations with fixed and dynamic LBCs, are regional dependent, causing up to 10–20% decrease over the western United States in future summertime $PM_{2.5}$ pollution. Therefore, it is important to consider the relative contributions of emissions scenarios, climate conditions, and LRT to the major $PM_{2.5}$ components in future U.S. air quality regulation.

1. Introduction

Particulate matter (PM) is a mixture of small particles and liquid droplets, suspending in the atmosphere as atmospheric aerosols. PM with aerodynamic diameter of 2.5 μm or less ($PM_{2.5}$) is one of the major air pollutants (EPA, 2004), causing negative effects on human health (e.g., Anderson, 2009; Schwartz et al., 2002), impairing visibility (e.g., Hyslop, 2009), and affecting climate (IPCC, 2007). $PM_{2.5}$ pollution is highly dependent on emissions and favorable weather patterns. Consequently, future climate change as well as emissions change could significantly influence $PM_{2.5}$ levels (Jacob and Winner, 2009). For instance, higher temperatures projected for the future can reduce nitrate aerosol concentrations through changing the ammonium and nitrate partitioning (Dawson et al., 2007); the shift of atmospheric circulation pattern under future climate can influence the distribution and concentrations of air pollution (Leibensperger et al., 2008). Vice versa, changes of $PM_{2.5}$ pollution can also affect the global radiative balance and future climate (Leibensperger et al., 2012a, b). As such, better understanding of potential changes of future $PM_{2.5}$ pollution is important to future air quality regulation and climate change adaptation strategies.

Global and regional chemical transport models (CTMs), when coupled with global climate models (GCMs), are widely used tools for studying the complex interactions between climate and PM pollution under future emissions and associated changes in climate (e.g., Avise et al., 2009; Heald et al., 2008; Liao et al., 2006; Pye et al., 2009;

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Racherla and Adams, 2006; Tagaris et al., 2007; Tai et al., 2012; Unger et al., 2006). Jacob and Winner (2009) reported a substantial range of uncertainty ($-0.1 \pm 1\ \mu g/m^3$), even in the sign of effects, across these studies on sensitivity of future $PM_{2.5}$ pollution to climate change. Recent studies, including Jiang et al. (2010) and Kelly et al. (2012), have not narrowed the uncertainty. Most of these studies use a single emissions scenario and/or climate projection (defined as climate state hereafter) (e.g., Liao et al., 2006; Pye et al., 2009; Tagaris et al., 2007), so effects of emissions change and climate change under multiple future scenarios are not evaluated in a consistent modeling system. The U.S. $PM_{2.5}$ pollution is also influenced by long-range transport (LRT) of air pollutants such as transpacific transport from Asia (e.g., Dunlea et al., 2009; Heald et al., 2006) and Mexico (Mukerjee et al., 2001). Recent development of regional CTM system has used semi-hemispheric Community Multiscale Air Quality (CMAQ) modeling to provide LBCs for the nested CMAQ with higher spatial resolution (Gonzalez-Abraham et al., 2015; Mathur et al., 2017). The consideration of multiple emissions scenarios, climate states, and LRT effects in a consistent modeling system is important to enhancing the understanding of future U.S. changes in $PM_{2.5}$ pollution.

This study investigates the $PM_{2.5}$ pollution in the continental United States under combinations of different emissions scenarios, climate states and LRT effects, to identify their relative contributions and responses of major $PM_{2.5}$ species. Section 2 presents the set-up of the modeling system and observation datasets used in this study. In section 3, the $PM_{2.5}$ simulations for present-day are evaluated relative to observations. Section 4 investigates projections of $PM_{2.5}$ pollution to examine the individual effects and estimate the response of major $PM_{2.5}$ species. Finally, we identify the major forcings affecting trends in future $PM_{2.5}$ pollution, and discuss associated uncertainties.

2. Modeling system and observations datasets

2.1. Model description

We used a regional modeling system that includes the U.S. Environmental Protection Agency (EPA) CMAQ model coupled with a regional climate model (RCM). He et al. (2016) used this system to simulate the present-day and future U.S. ozone pollution changes and quantify the relative contributions of major sources for projecting the changes. The present paper uses the same modeling system and simulations to examine influences of emissions, climate change, and long-range transport on ground-level $PM_{2.5}$ concentrations.

Global present-day (1995–1999) and future (2048–2052) climate states, used as boundary and initial conditions for the regional system, were from simulations of the National Center for Atmospheric Research (NCAR) Community Climate System Model version 3 (CCSM3) (Collins et al., 2006a, 2006b). The future climate projections were conducted under the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) A1B and A1Fi, respectively (Nakicenovic et al., 2000). Regional climate was downscaled using a regional climate model based on the 5th Pennsylvania State University/NCAR Mesoscale Model (CMM5) (Liang et al., 2001). CMM5 is a useful downscaling tool to improve GCM climate simulations (Liang et al., 2006). Several studies demonstrated that CMM5 has good performance of simulating present-day and future climate, especially precipitation, which provides high credibility in future climate projections (Liang et al., 2004a, 2004b; 2006, 2007, 2008).

To drive the regional CTM, present-day U.S. anthropogenic emissions were based on the 2002 EPA National Emissions Inventory (NEI 2002, available from http://www.epa.gov/ttnchie1/net/2002inventory.html), with the 1999 Mexico National Emissions Inventory (available from http://www.epa.gov/ttnchie1/net/mexico.html) and 2000 Canadian emissions inventory (available from http://www.epa.gov/ttnchie1/net/canada.html) as supplementary. Biogenic emissions and soil nitrogen oxide (NO) emissions were produced using the Biogenic Emissions Inventory System (BEIS, v3.13) built in the Sparse Matrix Operator Kernel Emissions model (SMOKE) based on the Biogenic Emissions Landuse Database data (BELD, available from http://www.epa.gov/ttnchie1/emch/biogenic/). Future anthropogenic emissions rates were estimated by applying scaling factors derived from the IPCC SRES report (Nakicenovic et al., 2000). The scaling factor for each inventoried pollutant in each region was calculated for A1B and A1Fi emissions scenarios of the years 2050. The U.S. and Canada were treated similarly to the Organization for Economic Cooperation and Development 1990 (OECD90) region defined in IPCC. Mexico was treated as the Africa, Latin America, and Middle East (ALM) region. It should be noted that the scaling factors for sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), and non-methane volatile organic compounds (NMVOCs) were based on the IPCC projected emissions. The scaling factors for PM_{10}, $PM_{2.5}$ and ammonia gas (NH_3) were derived from the IPCC population projections. Projected emissions for 2050 were calculated by applying those scaling factors to the 2002 United States National Emissions Inventory (NEI 2002), the 1999 Mexico National Emissions Inventory (MNEI), and the 2000 Canadian baseline inventory. The underlying assumption here, consistent with the IPCC projections, is that the numbers of sources do not change. Only the magnitudes of existing sources change, which implies that the geographic distribution of emissions remains the same. The A1B scenario emphasizes a balanced use of fossil fuel and renewable energy, while the A1Fi scenario emphasizes an intensive usage of fossil fuel. Therefore, A1B and A1Fi represent a relatively 'clean' outlook and a 'dirty' outlook respectively, compared with the present-day emissions. Emissions inputs for CMAQ were preprocessed using SMOKE v2.4 (Houyoux et al., 2000; UNC, 2007) driven by the CMM5 downscaled meteorology (Tao et al., 2007) and speciated for the Carbon Bond 5 (CB05) chemical mechanism (Yarwood et al., 2005).

To identify LRT effects through chemical LBCs on $PM_{2.5}$ simulations, we created two sets of LBCs. Fixed LBCs were predefined vertical profiles of species concentrations as a function of height (CMAS, 2007) which are built in CMAQ and widely used as the default chemical LBCs (e.g., Hogrefe et al., 2011). Dynamic LBCs were derived from the NCAR Community Atmospheric Model with Chemistry (CAM-Chem, v3) simulations driven by the same CCSM3 climate and emissions (see details in Lei et al., 2012, 2013). Hourly 3-D LBCs were generated with converting CAM-Chem outputs to CB05 species. Through comparing CMAQ simulations under fixed and dynamic LBCs, we can assess the uncertainty of LRT effects on the future U.S. $PM_{2.5}$ pollution.

CMAQ version 4.6 was used to conduct present-day and future simulations of air quality over the Contiguous United States (CONUS). The modeling domain has 195 × 138 grids with horizontal resolution of 30 km (Fig. 1a) and 35 vertical layers from surface to 50 hPa. Ten numerical experiments were conducted under different combinations of emissions, climate and LBCs (Table 1). Each case was run for 5 years (1995–1999 or 2048–2052), and due to limited computing resources, future cases with dynamic LBCs were only conducted for summer (June, July, and August).

2.2. Observation data and analysis method

To evaluate the model performance of CMAQ, data of ground-level $PM_{2.5}$ concentrations were obtained from the EPA Air Quality System (AQS) database (http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm), the Clean Air Status and Trends Network (CASTNET, http://epa.gov/castnet/javaweb/index.html), and the Interagency Monitoring of Protected Visual Environments network (IMPROVE, http://vista.cira.colostate.edu/improve/). $PM_{2.5}$ speciation data from CASTNET and IMPROVE were also used to evaluate the composition of present-day $PM_{2.5}$ simulations. The filter-based CASTNET technique provides mass concentrations of sulfate, nitrate, and ammonium, which are used to construct the total $PM_{2.5}$ mass of inorganic species (hereafter named $PM_{2.5,SNA}$). Since CASTNET and