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Modeling dry and wet deposition of sulfate, nitrate, and ammonium ions in Jiuzhaigou National Nature Reserve, China using a source-oriented CMAQ model: Part II. Emission sector and source region contributions

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HIGHLIGHTS

- First source-oriented modeling study on regional air pollution deposition.
- Industry and power plants are major contributors to SO_4^{2-} and NO_3^- deposition.
- Fertilizer and manure management are major sources of NH_4^+ deposition.
- >90% of SO_4^{2-} and NO_3^-, and >70% of NH_4^+ deposition are due to long range transport.

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ABSTRACT

A source-oriented Community Multiscale Air Quality (CMAQ) model driven by the meteorological fields generated by the Weather Research and Forecasting (WRF) model was used to study the dry and wet deposition of nitrate (NO_3^-), sulfate (SO_4^{2-}), and ammonium (NH_4^+) ions in the Jiuzhaigou National Nature Reserve (JNNR), China from June to August 2010 and to identify the contributions of different emission sectors and source regions that were responsible for the deposition fluxes. Contributions from power plants, industry, transportation, domestic, biogenic, windblown dust, open burning, fertilizer, and manure management sources to deposition fluxes in JNNR watershed and four EANET sites are determined. In JNNR, 96%, 82%, and 87% of the SO_4^{2-}, NO_3^- and NH_4^+ deposition fluxes are in the form of wet deposition of the corresponding aerosol species. Industry and power plants are the two major sources of SO_4^{2-} deposition flux, accounting for 86% of the total wet deposition of SO_4^{2-}, and industry has a higher contribution (56%) than that of power plants (30%). Power plants and industry are also the top sources that are responsible for NO_3^- wet deposition, and contributions from power plants (30%) are generally higher than those from industries (21%). The major sources of NH_4^+ wet deposition flux in JNNR are fertilizer (48%) and manure management (39%). Source-region apportionment confirms that SO_2 and NO_x emissions from local and two nearest counties do not have a significant impact on predicted wet deposition fluxes in JNNR, with contributions less than 10%. While local NH_3 emissions account for a higher fraction of the NH_4^+ deposition, approximately 70% of NH_4^+ wet deposition in JNNR originated from other source regions. This study demonstrates that S and N deposition in JNNR is mostly from long-range transport rather than from local emissions, and to protect JNNR, regional emission reduction controls are needed.

1. Introduction

Excessive deposition of sulfur (S) and nitrogen (N) threatens many protected areas such as national parks and nature reserves across the world (Bytnerowicz et al., 2007; Neufeld and Chappelka, 2007; Phoenix et al., 2006; Sullivan et al., 2011a,b). Close to one of the world's largest acid rain affected zones in southwestern China (Larssen et al.,

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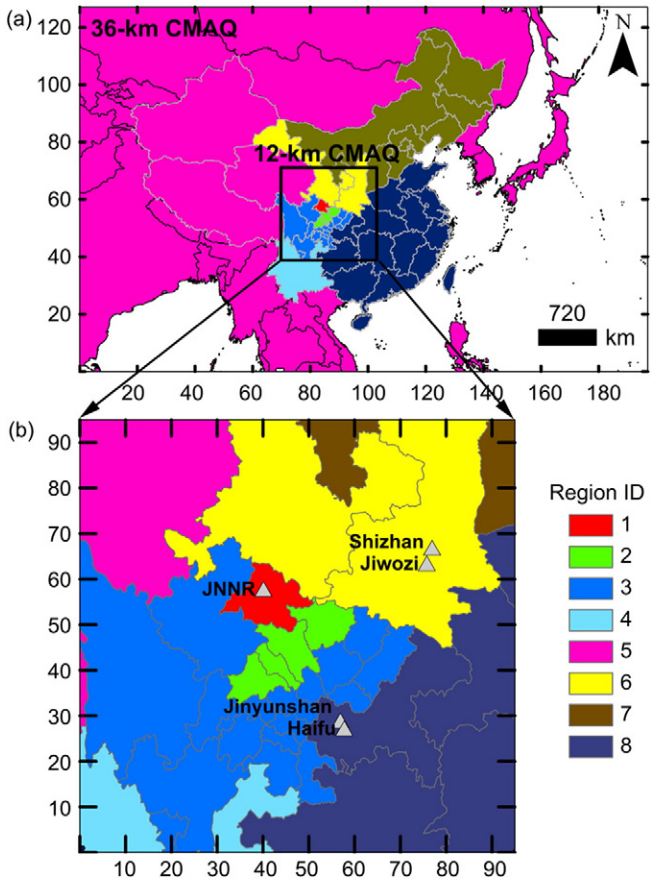

Fig. 1. CMAQ modeling domains and designation of source regions: 1—JNNR and three surrounding counties (Jiuzhaigou, Wen, and Pingwu counties); 2—metropolitan cities closest to JNNR in Sichuan Province (Chengdu, Mianyang, Deyang, and Guangyuan); 3—Other parts of Sichuan Province; 4—Yunnan; 5—West Provinces (Xinjiang, Qinghai, and Tibetan) and the other countries in the 36-km domain; 6—Northwest Provinces (Gansu and Shanxi Provinces); 7—North Provinces (Beijing, Tianjing, Hebei, Shanxisheng, Inner Mongolian, Ningxia, Liaoning, Jilin, and Helongjiang); and 8—Other Central, East, and South Provinces.

2006) (Fig. 1), Jiuzhaigou National Nature Reserve (JNNR) is one of the protected areas that receive elevated atmospheric inputs of S and N through wet and dry deposition (Qiao et al., 2014, 2015b). To develop relevant measures to protect JNNR and other vulnerable ecosystems, the sources of S and N deposition need to be determined.

Source attribution of ionic components in dry and wet deposition has been studied using a number of statistical source apportionment methods, including the Positive Matrix Factor (PMF) (Li et al., 2011; Qiao et al., 2015a) and Principal Component Analysis (PCA) (Huang et al., 2009; Zhou et al., 2012). For JNNR, a source apportionment study using PMF model determined that fossil fuel combustion and agricultural activities were major sources of S and N deposition (Qiao et al., 2015a). As anthropogenic precursor emissions were low in JNNR, most of the S and N deposited in JNNR were likely from other regions (Qiao et al., 2015a). However, statistical source apportionment analyses alone cannot determine the emission sector contributions to secondary particulate matter and the source regions that are responsible for dry and wet deposition in JNNR. The source and region contribution information is necessary to design effective emission control strategies for JNNR and other vulnerable ecosystems in southwestern China, one of the world's biodiversity hotspots (Conservation International 2014).

Previous applications of chemical transport models (CTMs) for dry and wet deposition analysis were mostly focused on determining the deposition fluxes on various types of land/water surfaces (Carmichael et al., 2002; Wang et al., 2004; Zhang et al., 2004). While source-oriented CTMs have been applied to determine emission sector and source region contributions to air pollutant concentrations (Chen et al., 2010; Ying and Kleeman, 2006, 2009; Zhang et al., 2012a), applications of CTMs for source apportionment of deposition fluxes have rarely been reported. Recently, Ge et al. (2014) applied a source-tracking CTM to determine the total deposition of S and N in nine different regions in China and East Asia, and input/output of S and N from each region to other regions. They determined that East and Central China were two primary export regions of S and N deposition for neighboring countries. However, they did not quantify the contributions from different emission sectors to S and N deposition.

In this study, a source-oriented Community Multiscale Air Quality (CMAQ) model (Ying et al., 2014a; Zhang et al., 2012b) was used to investigate the contributions of different sectors and regions to deposition of SO_4^{2-}, NO_3^-, and NH_4^+ in JNNR and surrounding areas from June to August 2010, which is part of the wet season (from late April to early October) and accounts for 40% of annual precipitation. To the best of the authors' knowledge, this is the first application of the source-oriented CMAQ model to study contributions of emission sectors and source regions to dry and wet deposition. A complete evaluation of the predicted SO_4^{2-}, NO_3^-, and NH_4^+ deposition against observations was presented in a companion paper (Qiao et al., 2015a). In this paper, the contributions of emission sectors and source regions to SO_4^{2-}, NO_3^-, and NH_4^+ deposition fluxes in JNNR are presented. The analyses elucidate the local vs. regional contributions to deposition fluxes, and emphasize the importance of regional emission controls in protecting vulnerable ecosystems.

2. Mo0064el description

A source-oriented version of the CMAQ model based on CMAQ 4.7.1, with a modified gas phase chemical mechanism SAPRC-99 (S99) (Carter, 2000) and aerosol module version 5 (AERO5), was used in this study. Details of the source-oriented CMAQ have been described by Zhang et al. (2012a) and Ying et al. (2014b). In summary, unlike CTMs that represent precursors and their oxidation products using generic species thus do not retain their emission sector and source region origin information, the source-oriented CMAQ model uses multiple tagged reactive species to represent the same species that originated from different emission sectors or source regions. For example, NO_2_S1 and NO_2_S2 can be used to represent NO_2 from two different emission sectors or source regions. The original S99 mechanisms are expanded to simulate the gas phase reactions of the source-tagged reactive species. For example, based on the original reaction $NO_2 + OH \rightarrow HNO_3$, two additional reactions can be added for the tagged species NO_2_S1 and NO_2_S2, which form HNO_3_S1 and HNO_3_S2, respectively. The aerosol and cloud modules are also modified to include additional aerosol nitrate species to correctly account for gas-to-particle partitioning of the tagged HNO_3 species. The additional gas and aerosol species would go through the same physical and chemical processes treated in the CMAQ model, including dry and wet deposition processes, as their non-tagged counterparts. Hourly deposition fields for all species are produced along with concentration fields, allowing direct source apportionment of the dry and wet deposition fluxes in all grid cells in the model domain. The current version of the source-oriented CMAQ model is capable of tracking emissions from nine emission sectors or source regions, with 304 gas phase species and ~2000 gas phase reactions.

3. Model application

The source-oriented CMAQ model is applied to study dry and wet deposition of SO_4^{2-}, NO_3^-, and NH_4^+ in JNNR and surrounding regions during a three-month period from June to August, 2010. Detailed model domain set up, meteorological simulations, and anthropogenic and biogenic emission processing have been described in Qiao et al. (2015a). In summary, a two-level nested domain with horizontal resolutions of 36-km and 12-km was used, with the 12-km domain (95×95