



Deposition of sulfur and nitrogen components in Louisiana in August, 2011

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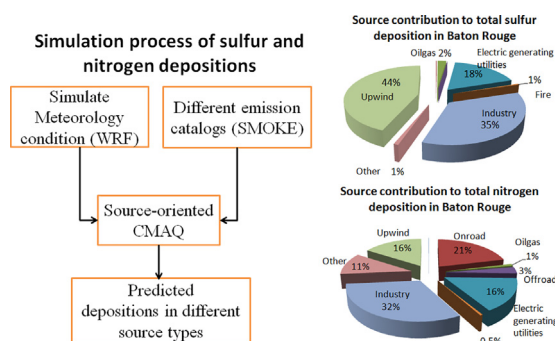
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HIGHLIGHTS

- Deposition of sulfur and nitrogen was not well understood in Louisiana.
- Source-oriented CMAQ tracks sulfur and nitrogen in all atmospheric processes.
- Aerosol phase sulfate is dominates sulfur deposition and EGU/industry/upwind sources are main sources.
- NO₂, HNO₃, NH₃, and aerosol phase nitrate are main nitrogen deposition forms. On-road, EGU, industry and agriculture are major sources for nitrogen deposition.

GRAPHICAL ABSTRACT



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ABSTRACT

Dry and wet deposition of sulfur and nitrogen contained air pollutants lead to increase of sulfur and nitrogen to the surface, causing acidification of surface water bodies and subsequent damage to aquatic and terrestrial ecosystems. Louisiana has abundant water resources and it is important to protect water resources from excessive atmospheric deposition of sulfur and nitrogen. However, the information for understanding sulfur and nitrogen deposition and adverse effects in Louisiana is limited. In this study, a source-oriented version of the Community Multi-scale Air Quality (CMAQ) model was used to simulate emission, formation, transport, and deposition of sulfur and nitrogen species in Louisiana in August 2011 to understand the forms and quantities of sulfur and nitrogen deposition due to wet and dry processes in Louisiana, to show the spatial and temporal variations of deposition fluxes, and to quantify the contributions of different sources to sulfur and nitrogen deposition. Aerosol phase sulfate (AeroSO₄) has the highest monthly total flux of 6 kg S/ha in wet deposition of sulfur. Major forms of sulfur dry depositions are AeroSO₄ (~1 kg S/ha) and SO₂ (~3 kg S/ha). Nitrogen deposition is mainly in forms of NO₂, HNO₃, NH₃, and aerosol phase nitrate (AeroNO₃). Electric generating utilities (EGU) are the largest contributor to sulfur depositions with monthly total flux of 1.6 kg S/ha, followed by industry (1 kg S/ha) and upwind sources. On-road vehicles and industry are important to nitrogen (except ammonia) depositions with maximum contributions of 0.6 kg N/ha and 0.8 kg N/ha, respectively. The dominate source of ammonia is “Other” implicit sector including agricultural activities.

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1. Introduction

Sulfur and nitrogen containing air pollutants are directly emitted from anthropogenic and biogenic sources or secondarily formed through complex reactions using the atmosphere as a potent oxidizing medium. Sulfur is mostly emitted as SO_2 from combustion sources and oxidized to sulfate. Nitrogen is emitted as oxides (NO_x) or ammonium (NH_3), and can exist in various forms including nitric acid in gas phase and nitrate and ammonium in aerosol phase. The ultimate pathways that remove sulfur and nitrogen species from the atmosphere are wet and dry depositions. Although deposition of these pollutants reduces their concentrations in atmosphere, it leads to the increase of sulfur and nitrogen fluxes to the earth surface. Atmospheric deposition of sulfur and nitrogen has been related to acidification of surface water bodies (lakes, rivers, and coasts) (Heard et al., 2014) and subsequent damages to aquatic ecosystems as well as forests and vegetation (Benedict et al., 2013; Bytnerowicz et al., 2007; Vinebrooke et al., 2014) as well as soil acidification (Monteith et al., 2007) since a majority of sulfur and nitrogen species are in acidic forms. Also, as an essential nutrient element for all livings, deposition of nitrogen causes nutrient imbalances of ecosystems, contributing to long-term eutrophication, increase of weedy plant species, and loss of native species (Bergström and Jansson, 2006).

Efforts had been made to understand and control atmospheric deposition of sulfur and nitrogen in the US and worldwide. The National Atmospheric Deposition Program (NADP) (Lamb and Bowersox, 2000) was initiated in 1977 to measure atmospheric deposition and the Clean Air Status and Trends Network (CASTNET) (Baumgardner et al., 2002) was established in 1991 to assess the trends in acidic deposition. Total sulfur deposition in the East US declined by 46% and nitrogen deposition decreased by 26% from 1990 through 2009 (MACTEC Engineering and Consulting, 2011).

Studies have also been conducted to understand the deposition of sulfur and nitrogen in the US and worldwide and related effects. Sun et al. (2016) found 86.64% of areas of the contiguous United States are dominated by the oxidized nitrogen deposition and Li et al. (2016) found dry deposition of ammonium increased in recent years and plays an especially key role in nitrogen deposition, contributing from 19% to 65% in different regions in US. It indicates that future progresses toward reducing US nitrogen deposition will be increasingly difficult without a reduction in ammonia emissions. Vet et al. (2014) carried out a global assessment of precipitation chemistry and deposition to address major gaps and uncertainties in global deposition measurements. McDonnell et al. (2014) evaluated potential long-term effects of atmospheric nitrogen deposition on subalpine ecosystems in Rocky Mountain National Park, Colorado. A global 3-D chemical transport model (GEOS-Chem) was applied to identify the sources and processes that

control nitrogen deposition to an ensemble of biodiversity hotspots worldwide in Paulot et al. (2013) and found ammonium (50%) and anthropogenic sources (50%), with important contributions from Asia and North America. Although previous works expanded our knowledge toward atmospheric deposition of sulfur and nitrogen in US, their results were limited by the lack of deposition observations and sources apportionment methods were not used to analyze the detailed potential anthropogenic sources.

Connecting the Mississippi river and the Gulf of Mexico, Louisiana has abundant water resources with approximately 11% of the total surface area composed of various water bodies. The state has >40,000 miles of rivers, streams and bayous, and 400 miles of coastline. Almost every aspect of economy in Louisiana can be tied to the development and utilization of water resources. Thus, it is important to protect water resources from excessive atmospheric deposition of sulfur and nitrogen. However, there are only two sites in NADP and no sites in CASTNET located in Louisiana. Thus, the information that can be obtained from the observation systems for understanding the deposition of sulfur and nitrogen and the adverse effects in Louisiana is very limited. Critical information is missing for designing effective control measures to reduce the deposition fluxes of sulfur and nitrogen, such as the forms in which they are deposited, the contributions of different source sectors, and the spatial patterns.

The climate in Louisiana is humid subtropical with long, hot and humid summer. Rain is frequent throughout the year and more typical during the summertime. Given the increased amount of rainfall and precipitation in summer, August was selected to represent the whole summer in this work.

Thus, this paper aims to 1) understand the forms and quantities of sulfur and nitrogen deposition from wet and dry processes in Louisiana; 2) pinpoint the spatial and temporal variations of sulfur and nitrogen fluxes; and 3) quantify the contributions to sulfur and nitrogen deposition from different source sectors.

2. Methodology

2.1. Model description

The EPA Community Multiscale Air Quality (CMAQ) v4.7.1 was used in this study with source-oriented techniques. The modified photochemical mechanism SAPRC-99 (Carter, 2000) was used with aerosol module version 5 (AERO5). The source-oriented techniques were utilized to track emissions of nitrogen and sulfur species from different emission sectors or source regions, and their corresponding final products in the gas or aerosol phases by modifying the photochemical mechanism SAPRC-99 and aerosol module version 5 (AERO5). The detailed

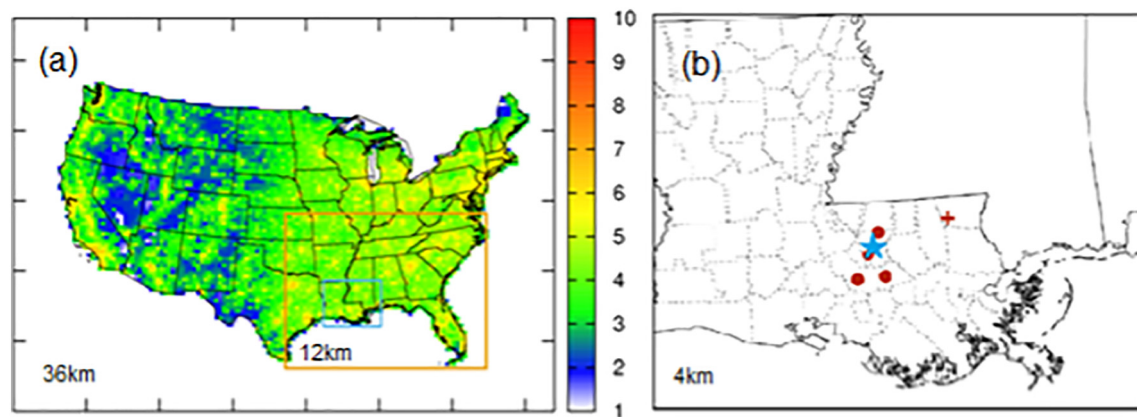


Fig. 1. (a) Setting of the nested domains with population density in log scale and (b) locations of the observation sites of deposition flux (red-cross), and $\text{PM}_{2.5}$ components (red dots) within the 4-km domain (Baton Rouge area is marked by blue star). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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