

Chapter 7

Long-Term Trends in Sulfur and Reactive Nitrogen Deposition Across the Northern Hemisphere and United States



Rohit Mathur, Yuqiang Zhang, Christian Hogrefe and Jia Xing

Abstract We model the changes in wet and dry deposition amounts of reactive nitrogen and sulfur over the 1990–2010 period using the WRF-CMAQ modeling system. WRF-CMAQ simulations for this 21-year period were conducted over a domain covering the northern hemisphere using a horizontal resolution of 108 km and a nested domain over the contiguous U.S. using a grid of 36 km resolution. The impacts of contrasting changes in emission patterns across the Northern Hemisphere, i.e., reductions in North America and Europe vs. increases across regions in Asia, on changing deposition amounts over terrestrial and aquatic ecosystems in these regions is analyzed.

7.1 Introduction

A detailed understanding of the distribution and fate of atmospheric sulfur (SO_x) and reactive nitrogen compounds (NO_y and NH_x) is desirable given their role in determining tropospheric acidic substances and particulate matter budgets and potential nutrient loading effects in sensitive ecosystems resulting from their atmospheric deposition. The ultimate fate of airborne SO_x , NO_y and NH_x is removal by wet scavenging and dry deposition, which in turn lead to a variety of environmental effects including altering net primary production, acidification, eutrophication and other nutrient loading effects. The gas-particle partitioning of airborne sulfur and reactive nitrogen regulates their transport distances since dry deposition velocity for fine particles is relatively low, and consequently their primary atmospheric sink is wet scavenging. Changing emissions patterns of NO_x , SO_2 , and NH_3 have likely altered both their atmospheric transport distances as well as deposition patterns and amounts.

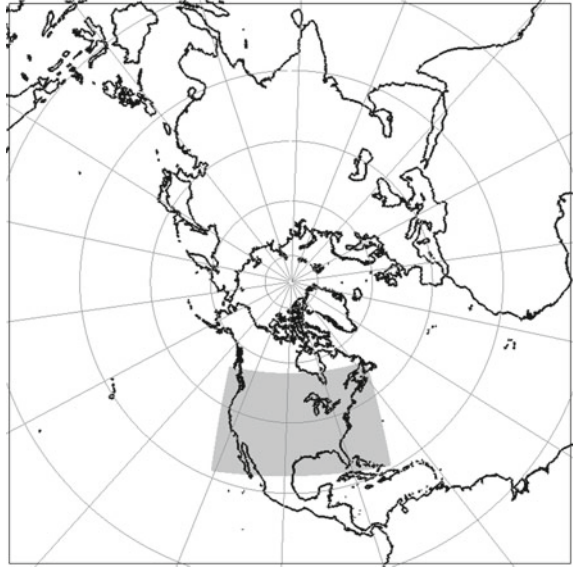
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Fig. 7.1 The Northern Hemisphere (outer) and nested Continental U.S. modeling domain (shaded)



7.2 Model Setup

The WRF-CMAQ modeling system was used to simulate the tropospheric composition and wet and dry deposition of various atmospheric species over a 21-year period spanning 1990–2010. Model simulations were performed over a domain encompassing the entire Northern Hemisphere (Fig. 7.1), set on a polar stereographic projection and discretized with a horizontal grid spacing of 108 km [1]. Space and time varying lateral boundary conditions from these large scale simulations were then provided to a 36 km resolution simulation for the same period over the Continental U.S. (CONUS) domain (shaded region in Fig. 7.1). Year specific emissions for the Northern Hemispheric domain were derived from the EDGARv4.2 global emission inventory [2] while those for the CONUS domain were based on the inventory of Xing et al. [3].

Hourly dry and wet deposition amounts from the WRF-CMAQ simulations were aggregated to seasonal and annual totals for each of the 21 years and their spatial distributions and temporal trends were analyzed.

7.3 Results and Discussion

Model estimated 1990–2010 trends in annual total deposition (wet + dry) of sulfur and inorganic nitrogen (IN) across the Northern Hemisphere are shown in Fig. 7.2. The directionality, magnitude and spatial variability of the model estimated wet deposition trends for sulfur and IN were found to be in reasonable agreement with

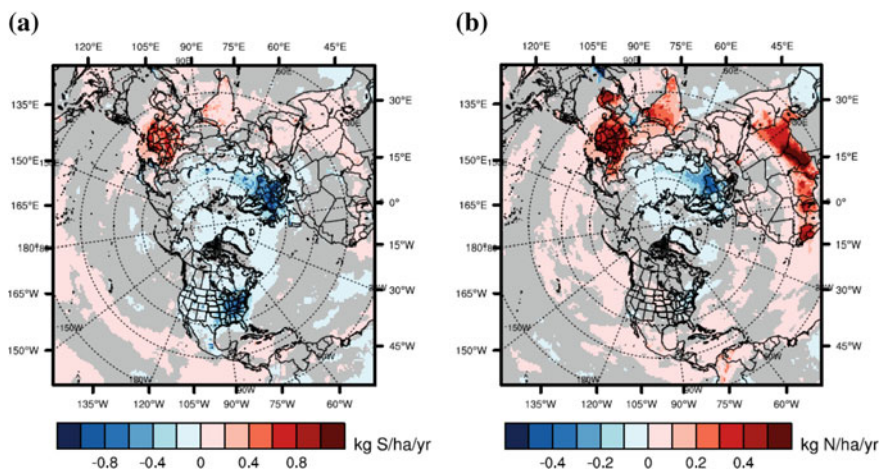


Fig. 7.2 1990–2010 trends in annual total wet + dry deposition across the Northern Hemisphere for **a** total sulfur ($\text{SO}_2 + \text{SO}_4^{2-}$) expressed in kg S/ha/yr and **b** total inorganic nitrogen ($\text{NO}_y + \text{NH}_x$) expressed in kg N/ha/yr . Grey shaded areas represent locations with p -value > 0.05 for the standard two-tailed Student T-test, i.e., areas where the modeled trend estimates were not significant at the 95% confidence level

those inferred from NADP measurements in the U.S. and EMEP measurements in Europe (not shown). Strong decreasing trends in atmospheric sulfur deposition are noted across the continental U.S., western Europe, and regions of the north Atlantic Ocean influenced by continental outflow, in response to the significant reductions in SO_2 emissions over the past two decades. In contrast, rising SO_2 emissions during this period resulted in increasing sulfur deposition trends across large parts of Asia as well as large portions of the Pacific influenced by Asian outflow. The spatial distributions of the trends in IN deposition are influenced by trends in both NO_x and NH_3 emissions. In Europe where emissions of both species have seen reductions during this period, we note reductions in IN deposition with contrasting increases in regional deposition across Asia in response to growth in emissions of both NO_x and NH_3 .

The IN deposition across the U.S. however depicts decreasing trends in the north-eastern U.S. and outflow over the Atlantic but increasing trends in the mid-western states. This is further explained by trends in atmospheric deposition of oxidized and reduced nitrogen which are illustrated in Fig. 7.3. Reductions in NO_x emissions resulted in systematic decreases in oxidized nitrogen with larger decreasing rates in the eastern than the western U.S. The increasing IN deposition trends over the east and central states were caused by NH_x deposition increases which in turn arise from increasing NH_3 emissions in these regions over the past two decades.

As illustrated in Fig. 7.4, strong increasing trends in dry deposition amounts of NH_x across the U.S. occurred during the 1990–2010 period and likely arise both from increasing NH_3 emissions but also perhaps from reduced transport distances. Reductions in SO_2 and NO_x emissions and their oxidation products have likely

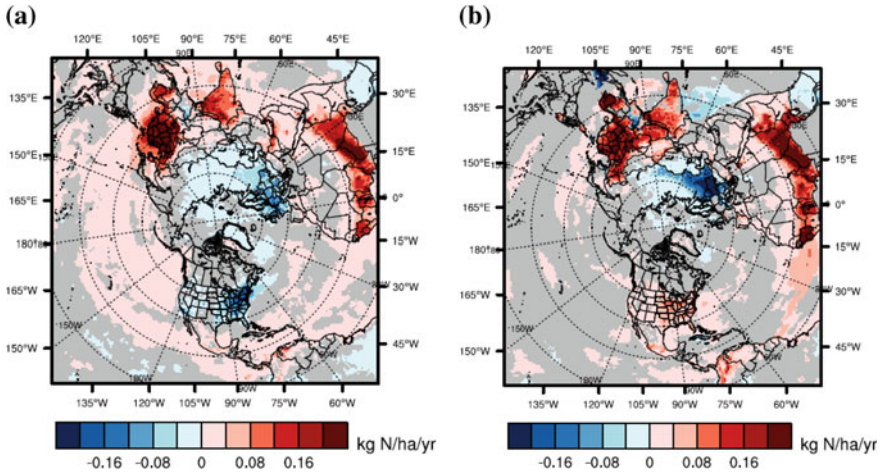
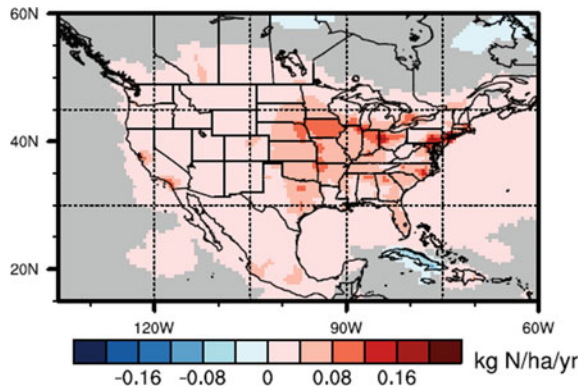


Fig. 7.3 1990–2010 trends in annual total wet + dry deposition (in kg N/ha/yr) across the Northern Hemisphere for **a** oxidized nitrogen (NO_y), and **b** reduced nitrogen (NH_x)

Fig. 7.4 1990–2010 trends in annual reduced nitrogen (NH_x) dry deposition (in kg N/ha/yr)



decreased the amounts of NH_x partitioning to the aerosol phase where scavenging by rain is the primary sink. Consequently, more NH_x remains in the gas-phase and dry deposits closer to the source regions as suggested in Fig. 7.4.

QUESTIONER: Talat Odman

QUESTION: The N-critical load in the northeast has decreased while it remained the same (or increased) in eastern North Carolina. What is this telling us in terms of sources of N?

ANSWER: The trends in total-N deposition are influenced by individual trends in reduced and oxidized-N deposition. In the northeast U.S., the 1990–2010 trends in N-deposition are dominated by trends in oxidized-N deposition while those in eastern North Carolina are dominated by trends in reduced-N deposition. Consequently, reductions in NO_x emissions across the northeastern U.S. have resulted in decreasing trends in N-deposition in the region. In contrast, increasing NH₃ emissions in eastern North Carolina resulted in increase in reduced-N and total-N deposition in that region.

QUESTIONER: Camilla Geels

QUESTION: Did you consider changes in land-use—could be important for dry deposition. If not, do you have any idea how significant they are?

ANSWER: To our knowledge no single dataset exists that describes in a consistent manner the changes in land-use for the multi-decadal period analyzed here. To minimize uncertainties associated with using different land-use data sets for different periods during 1990–2010, we decided to use the same land-use characterization for the entire period. While we acknowledge the likely impact of changing land use on simulated atmospheric dynamics and dry deposition amounts, we do not have a detailed assessment of the magnitude of the impact for this period. However, one could speculate that since both NH₃ and HNO₃ dry deposit efficiently to most surfaces, and since changes in land-use have been gradual, it is likely that the assumption of time invariant land-use does not have a significant impact on our estimated trends and results. Nevertheless, sensitivity simulations would need to be conducted to confirm this in a more quantitative manner.

Disclaimer The views expressed in this paper are those of the authors and do not necessarily reflect the views and policies of U.S. Environmental Protection Agency.

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