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Simulated effects of climate change on summertime nitrogen deposition in the eastern US

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Abstract

It is anticipated that climate change may impact regional-scale air quality and atmospheric deposition in the coming decades. To simulate the effects of climate change on nitrogen (N) deposition across numerous watersheds in the eastern US, we applied the NASA Goddard Institute for Space Studies General Circulation Model (GISS-GCM), Fifth Generation Pennsylvania State University/National Center for Atmospheric Research Mesoscale Model (MM5), Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system, and the US Environmental Protection Agency Community Multiscale Air Quality (CMAQ) Model. Keeping chemical initial and boundary conditions, land use, and anthropogenic area and point source emissions fixed, this modeling system was applied over five summers (June–August) from 1993 to 1997 and five summers from 2053 to 2057. Over these eastern US watersheds, the modeling system estimated 3–14% increases in summertime N deposition as a result of climate change. This increase is primarily due to the direct effects of climate change on atmospheric conditions and chemistry. Wet N deposition is predicted to increase as a result of increased precipitation, while dry N deposition is predicted to increase as higher surface temperatures favor gas-phase nitric acid to particulate nitrate. The simulated increase suggests that additional reductions in N oxides and/or ammonia may be needed to fully realize the anticipated benefits of planned reduction strategies, including the Clean Air Interstate Rule (CAIR).
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1. Introduction

The environmental impacts of acidic deposition, resulting primarily from emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x), have been studied for decades. It is well known that parts of the northeastern US, characterized by low buffering capacity, are particularly susceptible to the effects of deposition of sulfate (SO_4) and nitrate (NO_3). These effects include surface water acidification (e.g. Driscoll et al., 1998), depletion of base cations, and leaching/mobilization of aluminum in soils (e.g.

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Driscoll et al., 2001, 2003a), and poor growth or increased mortality of various tree species (e.g. Driscoll et al., 2001, 2003a). Excessive loading of nitrogen (N) and other nutrients to coastal estuaries is also known to contribute to increased eutrophication in these ecosystems (e.g. Jaworski et al., 1997).

It is thought that N deposition has increased by a factor of 3–4 globally since the mid-19th century, as a result of additional N releases associated with increases in food production and fossil fuel combustion (e.g. Galloway et al., 2004). Recent studies suggest that reductions in SO_2 emissions have led to declining SO_4 deposition over the eastern US. Through the 1980s and 1990s, N deposition did not change much, since changes in emissions of NO_x and ammonia (NH_3) were generally not as substantial (e.g. Lynch et al., 2000; Driscoll et al., 2001, 2003a; Baumgardner et al., 2002). However, recent analyses that include data after 2000 (e.g. Sickles and Shadwick, 2007; Lehmann et al., 2005) have found that wet and dry NO_3 deposition have decreased by about 10–25% in the northeastern US, possibly in response to more recent NO_x reductions (e.g. Phase II of Title IV of the Clean Air Act Amendments of 1990), while wet ammonium (NH_4) deposition in the northeastern US has not changed significantly. On the other hand, wet NH_4 deposition has increased by more than 25% over large parts of the western, central, and southeastern US (Lehmann et al., 2005).

Atmospheric deposition is a key source of N to watersheds in the eastern US. For example, Valigura et al. (1996) found that atmospheric deposition accounts for ~5–44% of the total N loading to bays and estuaries along the Atlantic coast. Here we apply a regional meteorological and air quality modeling system to estimate potential changes in N deposition as a result of climate change. We focus our analysis on a number of watersheds (or groups of watersheds) in the eastern US that are affected to some degree by atmospheric deposition, namely (1) Great Lakes; (2) Mobile Bay; (3) Albemarle/Pamlico Sounds; (4) Chesapeake Bay; (5) Delaware Inland Bays, Maryland Coastal Bays, and Barnegat Bay; (6) New York/New Jersey Harbor; (7) Long Island Sound and Peconic Bay; (8) Massachusetts Bays, New Hampshire Estuaries, Narragansett Bay, and Buzzard's Bay; and (9) Casco Bay (see the shaded regions in Fig. 1).

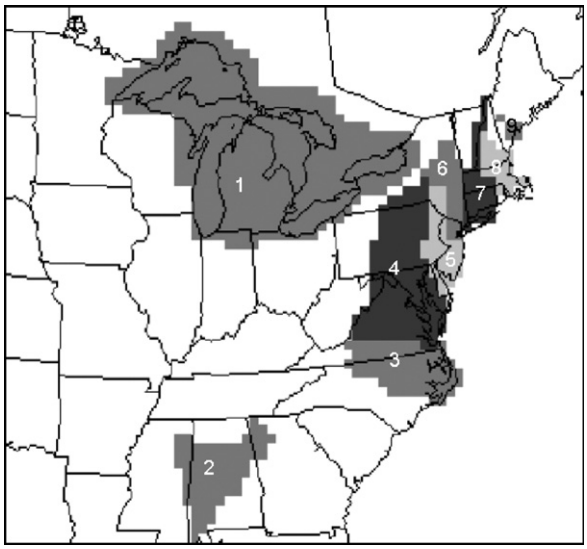

Fig. 1. The CMAQ 36 km modeling domain, with shaded regions denoting the approximate outlines of nine eastern watersheds (or groups of watersheds) labeled by number: (1) Great Lakes; (2) Mobile Bay; (3) Albemarle/Pamlico Sounds; (4) Chesapeake Bay; (5) Delaware Inland Bays, Maryland Coastal Bays, and Barnegat Bay; (6) New York/New Jersey Harbor; (7) Long Island Sound and Peconic Bay; (8) Massachusetts Bays, New Hampshire Estuaries, Narragansett Bay, and Buzzard's Bay; and (9) Casco Bay.

2. Overview of the modeling system and observational database

2.1. Modeling system

Estimates of near-current (1990s) and future (2050s) summertime surface N deposition fields were generated with a modeling system consisting of the Goddard Institute for Space Studies General Circulation Model (GISS-GCM; Russell et al., 1995); Fifth Generation Pennsylvania State University/National Center for Atmospheric Research Mesoscale Model (MM5; Dudhia, 1993); Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system (Houyoux et al., 2000); and the US Environmental Protection Agency (EPA) Community Multiscale Air Quality (CMAQ) Model (Byun and Schere, 2006). An overview of the modeling system follows, and the reader is referred to Hogrefe et al. (2004a, b) and Lynn et al. (2004) for additional details of the study design and model simulations.

Multi-decadal GISS-GCM simulations were performed from the 1990s through the 2080s, with future years consistent with the Intergovernmental

Panel on Climate Change Special Report on Emission Scenarios (IPCC SRES; please see http://www.ipcc.ch/ipccreports/sres/emission/index.htm) "A2" marker scenario. Very briefly, the A2 scenario predicts medium range increases in global greenhouse gas emissions (e.g. CO_2 increases by ~50%) and population growth by the 2050s. The $4^\circ \times 5^\circ$ GISS-GCM was coupled to the MM5 108 km outer domain in a one-way mode through the lateral boundaries and initial conditions. The nested 36 km MM5 domain included much of the eastern US (see Fig. 1), and the model simulations covered five summer seasons (June–August) for a recent period—1993–1997—and a future period—2053–2057. Key MM5 physics options used in this study include the Medium Range Forecast Planetary Boundary Layer Model (Hong and Pan, 1996), Grell cumulus parameterization (Grell, 1993), and the Rapid Radiation Transfer Model radiation scheme (Mlawer et al., 1997). It should be noted that the IPCC A2 scenario predicts substantial urban growth by the middle of the 21st century. For example, if the current land cover trends continue the amount of developed land in the Chesapeake Bay watershed may increase by about 80% by 2030 (Goetz et al., 2004), while developed land cover in the New York City metropolitan area—which affects several eastern US estuaries—may increase by more than threefold by 2050 (Solecki and Oliveri, 2004). Here we attempted to assess the effects of climate change alone without the potential concomitant changes in urban land cover and, hence, did not alter the MM5 land use classifications, to be consistent with the projected urban growth. We acknowledge that this is a limitation of the study and that land cover can certainly affect near-surface meteorological and air quality fields in regional-scale modeling (e.g. Civerolo et al., 2007; Ge et al., 2007).

Using SMOKE, we processed the county-level US EPA 1996 National Emissions Trends (NET96) Inventory to generate hourly, gridded anthropogenic emissions for this study. The area and point source emissions were identical for the two sets of simulations. The corresponding mobile source emissions were estimated with the US EPA Mobile5b Model (US EPA, 1994) using constant fleet composition and vehicle miles traveled (VMT) information while accounting for the effect of temperature on evaporative emissions. The biogenic emissions were created with the Biogenic Emissions Inventory System Version 2 (BEIS2; Geron et al., 1994; Williams et al., 1992), taking into account the effects of meteorology on these emissions. As a result, emissions of NO_x and NH_3 from the mobile sector and biogenic sources do depend weakly on temperature and show a slight increase of less than 0.5% in the future climate simulations. This represents a small indirect effect of climate change on atmospheric concentrations through the modification of pollutant emissions.

The air quality and deposition modeling was performed with CMAQ for the same two sets of summer periods, and the extent of the model domain is shown in Fig. 1. The model domain consists of 68×59 horizontal grid cells, with 16 vertical terrain-following σ_p levels; the lowest model layer is ~70 m thick. Below ~3 km, CMAQ and MM5 used identical vertical levels. Gas-phase chemistry was simulated with the Carbon Bond IV Mechanism (Gery et al., 1989). The initial and lateral boundary concentrations of trace gas and aerosol species were identical for the two sets of simulations. Ozone concentrations simulated by this modeling system for the summers of 1993–1997 were evaluated against observations by Hogrefe et al. (2004a).

2.2. Observational database

To perform a first-order assessment of the model-predicted deposition fields from the summers of 1993–1997, we compared the average 3-month accumulated wet and dry deposition amounts from CMAQ with the corresponding observed values. Monthly wet deposition of NH_4 and NO_3 were obtained from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN; http://nadp.sws.uiuc.edu). The NTN is a nationwide acid deposition network operated by a consortium of federal, state, and local governments, as well as groups representing Native American tribes, universities, industry, and other research entities. The NTN has been collecting wet deposition on a weekly basis since 1978 to assess temporal and spatial trends. We also obtained the daily and event-based wet deposition data from the Atmospheric Integrated Research Monitoring Network (AIRMoN-wet). AIRMoN-wet is sponsored by the National Oceanic and Atmospheric Administration/Air Resources Laboratory (NOAA/ARL; details can be found at http://www.arl.noaa.gov/research/programs/airmon.html), and these data are also available from the NADP web site (http://nadp.sws.

uiuc.edu). AIRMoN-wet has a much finer temporal resolution than the NTN, and has been operating as a sub-NADP network since 1992. For this analysis we calculated the 5-year average summer season accumulated wet NH_4 and NO_3 deposition amounts from 78 NTN sites and 7 AIRMoN-wet sites across the eastern US.

While there is ample information on wet deposition in the US, dry deposition is much more difficult to measure. We obtained the data from two inferential dry deposition networks, namely the US EPA Clean Air Status and Trends Network (CASTNet; http://www.epa.gov/castnet) and the dry deposition side of AIRMoN (AIRMoN-dry; http://www.arl.noaa.gov/research/programs/airmon.html). Dry deposition rates are computed from weekly average air concentrations coupled with detailed micrometeorological and ground surface information. Both inferential networks were operating during the 1990s, and we are not aware of systematic, direct, and long-term measurements of dry deposition with adequate spatial coverage to compare with the model predictions on a multi-year basis. CASTNet reports particulate NH_4 (pNH_4) and NO_3, as well as gas-phase nitric acid (HNO_3), while only NO_3 and HNO_3 are available from the AIRMoN data page. Dry deposition of gas-phase NH_3 is not reported by either network. Similar to wet deposition, we calculated the 5-year average summer season accumulated dry total nitrate (HNO_3+NO_3) and NH_4 deposition amounts from 27 CASTNet sites and 11 AIRMoN-dry sites (total NO_3 only). It is known that filter-based measurement of particulate NO_3 (pNO_3) and NH_4 may be confounded by volatilization of NH_4NO_3 (e.g. Ames and Malm, 2001; Hicks et al., 2001). It has been shown (e.g. Clarke et al., 1997; Dasch et al., 1989) that there is substantial uncertainty in the filter pack measurements of HNO_3 and NO_3—up to 30% for concentrations and 40% or more for inferred deposition velocities—such that dry deposition may only be accurate to within a factor of two. However, we used the CASTNet and AIRMoN-dry data "as is" with no corrections, but note that this may affect the model comparisons.

3. Results

3.1. Evaluation of the 1993–1997 simulations

Fig. 2(a) displays the mean, median, and interquartile range of summer season wet deposition of NH_4 as N (NH_4-N), NO_3 as N (NO_3-N), and the sum of NO_3-N and NH_4-N across the 85 NTN and AIRMoN-wet sites. Fig. 2(a) also includes the same information for precipitation at these 85 sites. Fig. 2(b) displays the same information for dry deposition at the CASTNet and AIRMoN-dry sites. Note that in Fig. 2(b), the statistics for dry NO_3-N deposition are included for all 38 sites and just the 27 CASTNet sites, since dry NH_4-N deposition data are not available from the AIRMoN-dry website. The CMAQ predictions were extracted from the model grid that contained the location of each monitor, and include gas-phase HNO_3. In terms of wet deposition, CMAQ tended to overpredict NH_4-N by ~54% and underpredict NO_3-N by ~28%; and NO_3+NH_4 was overpredicted by ~7% on average. Model predictions of seasonal average total N wet deposition were within a factor of two at each of the 85 wet deposition sites, with the exception of NTN site NY68 (Biscuit Brook, NY), a site in the Catskill Mountains where the model prediction was about a factor of three higher than observed. The ability of CMAQ to predict summertime wet deposition is often limited by the ability of the meteorological model to accurately predict convective rainfall (e.g. Morris et al., 2005; Davis and Swall, 2006). Note from Fig. 2(a) that the model tends to overpredict precipitation amounts at these sites by about 21% on average. At the NY68 site, modeled precipitation was more than twice that observed. Overall, the magnitudes of these errors are comparable to the results presented by Appel et al. (2005), who compared wet deposition of NO_3 and NH_4 simulated by CMAQ for the summer of 2001 against NADP measurements. While further CMAQ development is certainly needed to improve these estimates, this indicates that the modeling system applied in this study represents the current state of science in calculating climatological wet deposition estimates.

In terms of dry deposition, on average, CMAQ tended to underpredict NH_4-N by ~9% across the CASTNet sites; overpredict NO_3-N by ~71% across just the CASTNet sites, and ~57% across all sites; and NO_3+NH_4 by ~54% across the CASTNet sites. The scatter in NO_3+NH_4 dry deposition is somewhat larger than with wet deposition, with the ratio of predicted to observed N deposition at the CASTNet sites ranging from 0.84 to 3.85. However, given the known difficulties in measuring NO_3/HNO_3 on filter packs, and that

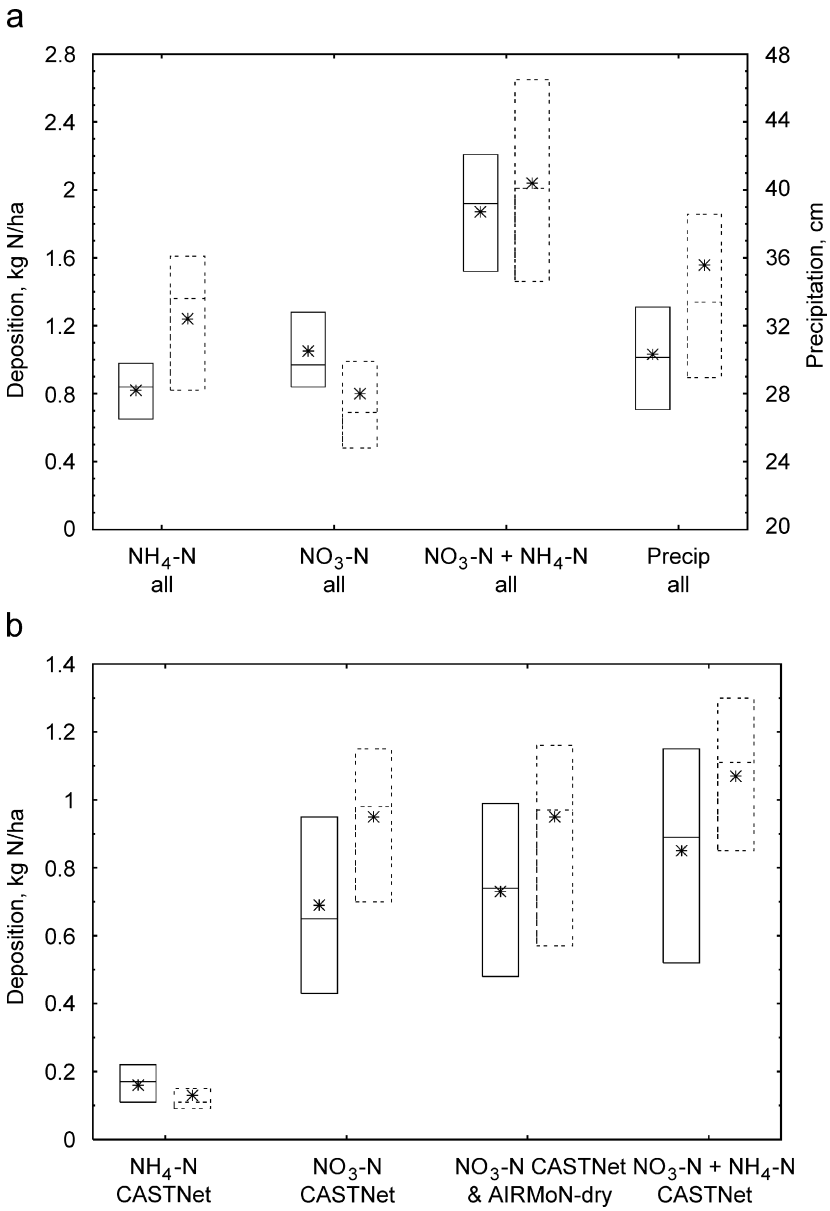

Fig. 2. Summary of observed (solid lines) and predicted (broken lines) summer season N deposition across all observational sites. The boxes denote the 25th percentile, median, and 75th percentile across all sites, the asterixes denote the averages. (a) Wet deposition and precipitation, 78 NTN, and 7 AIRMoN-wet sites, and (b) dry deposition, 27 CASTNet, and 11 AIRMoN-dry sites. For dry NO_3-N deposition, results are shown for CASTNet only and for both CASTNet and AIRMoN-dry sites.

CASTNet and AIRMoN-dry deposition values are inferred rather than directly measured, the model appears to perform satisfactorily in reproducing dry deposition of N compounds on a seasonal average basis. In particular, the model values are generally within a factor of two, the assumed uncertainty in inferential dry deposition measurements discussed in Section 2.2.

3.2. Comparison of future and near-current N deposition

Figs. 3(a) and (b) display the average summertime wet and dry N deposition, respectively, over the various watersheds in the eastern US for the 1990s and 2050s. Unlike the comparison with observations in the previous section, NH_3 is included in the

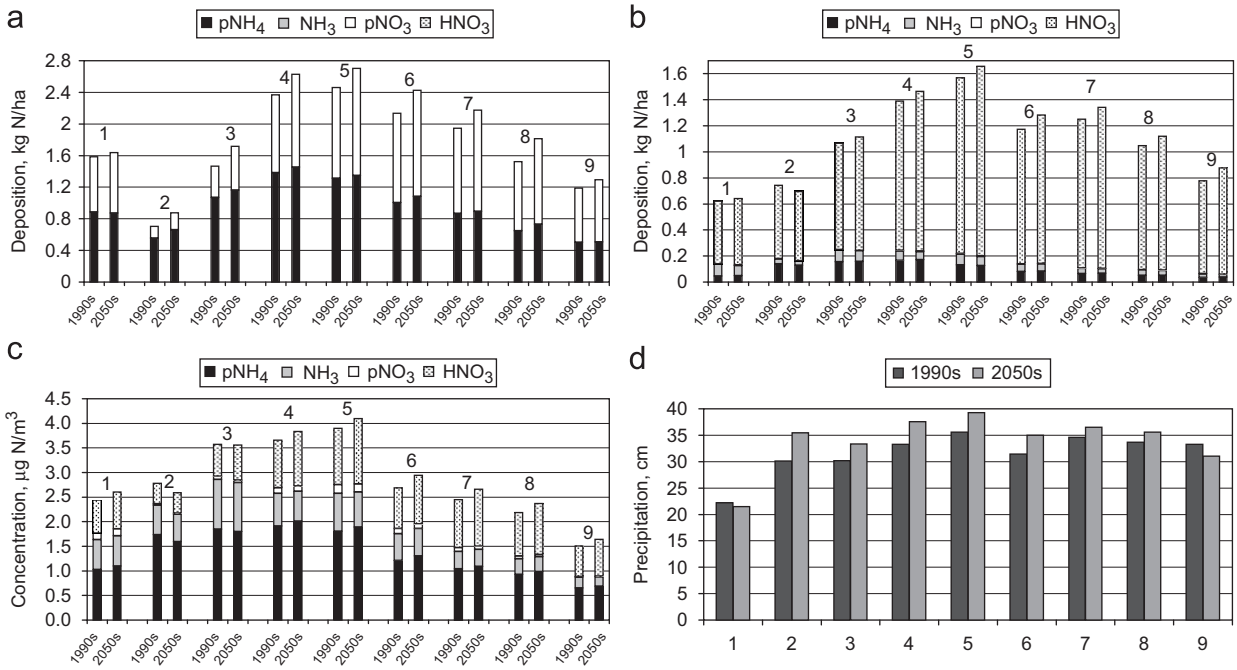

Fig. 3. Predicted N deposition, air concentrations, and precipitation over the various eastern watersheds during the summer seasons of the 1990s and 2050s. Watersheds are denoted by number. (a) Wet deposition (kg N ha^{-1}), (b) dry deposition (kg N ha^{-1}), (c) air concentrations (μg N m^{-3}), and (d) precipitation (cm).

model predictions in this section. To help interpret the changes in wet and dry deposition over these regions, we also computed the summer season air concentrations of these N species (Fig. 3(c)) and precipitation (Fig. 3(d)) for the two time periods. Over these watersheds, wet deposition accounted for ~49–71% of the total N deposition during the 1990s summer months. Wet deposition accounted for a slightly higher percentage (~56–72%) during the summer months of the 2050s.

Fig. 3(a) shows that gas-phase NH_3 and HNO_3 contribute negligibly to wet N deposition. During the 1990s, pNO_3 accounted for as little as 21% to total wet N deposition at Mobile Bay, to as much as 58% at the New England watersheds. This increased slightly to 25–61% during the 2050s. Wet N deposition increased over each watershed by ~3–25%. On a percentage basis, the smallest increases in wet N deposition occurred over the Great Lakes region (~3%) and Casco Bay (~9%). These two watersheds actually exhibited decreases in average summertime precipitation, by ~3% and 7%, respectively. The other seven watersheds exhibited increases in wet N deposition by ~10–25% and increases in precipitation by ~6–18%. Wet N deposition increased over each watershed despite the fact that air concentrations of pNO_3 decreased by ~2–21%, and changes in air concentrations of pNH_4 were relatively small (~8% decrease over Mobile Bay to ~7% increase over New York/New Jersey Harbor). Hence, it appears that increases in wet N deposition are largely dictated by increased precipitation amounts, which more than offset the effect of decreased pNO_3 concentrations resulting from warmer temperatures in the future climate scenario.

Fig. 3(b) indicates that dry N deposition is dominated by HNO_3, which accounts for 76–92% of dry N deposition in the 1990s and 77–93% in the 2050s. Only over Mobile Bay did dry HNO_3-N deposition decrease in the 2050s (0.56–0.53 kg N ha^{-1}); air concentrations of HNO_3 also decreased slightly over Mobile Bay (0.42–0.41 μg N m^{-3}). Dry deposition and air concentrations of HNO_3 increased over the other eight watersheds (deposition ~6–14%, concentrations ~10–22%), and total dry N deposition increased by ~3–13% over these watersheds. Temperatures across the model domain on average increased by ~1.5–2.5 °C in the future simulations, which led to a general shift from pNO_3 to gas-phase HNO_3. Deposition velocities of HNO_3 are generally

a factor of two larger than the corresponding deposition velocities of pNO_3 (e.g. Huebert et al., 1988; Peters and Bruckner-Schatt, 1995). At the same time, changes in air concentrations of NH_3 and pNH_4 were smaller than those in HNO_3; hence it appears that the changes in dry deposition are dominated by the increase in HNO_3 deposition over these watersheds.

Total N deposition, then, increased by about 3–14% across these watersheds in the simulation driven by the future regional climate fields under the A2 greenhouse gas scenario for the 2050s. This increase is primarily due to the direct effects of climate change on atmospheric conditions (increased temperatures and precipitation) and their effects on chemistry and gas/particle partitioning, since the point and area source emissions of NO_x and NH_3 were fixed and the associated increases in emissions in mobile and biogenic emissions were very small. This is of concern since it is thought that forested areas of the northeastern US are approaching N saturation (e.g. Driscoll et al., 2003b; Galloway et al., 2003), leading to further leaching of base cations and acidification of soils and surface waters, as well as reduced productivity and species richness in these forested regions. Anthropogenic emissions of NO_x are expected to decrease by about 45% between 2001 and 2020 over the Eastern US as a result of various emission control programs, including the Clean Air Interstate Rule (CAIR). This overall reduction is driven by reductions of about 70% from power plants and on-road mobile sources, with smaller reductions from non-road mobile sources and increases of about 20% from area and other point sources (US EPA, Clean Air Interstate Rule Emissions Inventory Technical Support Document, March 4, 2005, http://www.epa.gov/cair/pdfs/finaltech01.pdf). The results presented in this study indicate that in the long term some of the expected benefits from these emission reductions on N deposition may be counteracted by the effects of climate change, possibly requiring additional emission controls to achieve the desired reductions in deposition levels.

The results shown above illustrate the value of a coupled modeling system that can be used to estimate potential impacts on climate change on deposition; however, there are several limitations that should be mentioned here. First, this study only considers one in a range of possible future regional climate scenarios. Furthermore, choices made in the downscaling of global climate models to the regional scale can affect the simulated regional-scale temperature and precipitation patterns (e.g. Lynn et al., 2004; Liang et al., 2006) that could alter the simulated spatial patterns and magnitudes of deposition changes. In addition, the model simulations presented here only cover the three summer months. N deposition is a year-round issue, and it is important to characterize deposition during the winter months when nitrifying and denitrifying bacteria are less active and vegetation is less likely to take up available N. We also did not explore potential climate impacts on sulfur deposition in this paper, although SO_4 contributes substantially to the acidic deposition loading across much of the eastern US. Nor did we account for increased urban land cover which is expected over the coming decades in this region, or for changes in anthropogenic emissions related to technology development and the implementation of control measures. Finally, we focused primarily on the Great Lakes, Atlantic seaboard, and Gulf coast regions; important NH_4 and NO_3 source regions such as the Ohio River Valley and Southeastern US were not included here.

4. Summary

A modeling system has been applied to estimate summertime N deposition across important watersheds in the eastern US for a near-current (mid-1990s) and future (mid-2050s) period. The primary difference between these two sets of simulations was that regional climate change affected the future simulation. The modeling system's ability to reproduce observed N deposition during five summers of the 1990s is comparable to other studies, although part of the agreement between CMAQ and observations likely is due to compensating effects, i.e. overestimating NH_4 wet deposition and underestimating NO_3 wet deposition. In the simulations using the A2 regional climate fields for the 2050s, wet N deposition is predicted to increase, primarily as a result of higher precipitation, while dry N deposition is predicted to increase as a result of higher temperatures, leading to a shift from pNO_3 to HNO_3. In terms of total summertime N deposition, the effects of climate change are projected to cause an increase across eastern US watersheds by 3–14%, which may offset some of the benefits of planned emission reduction strategies. Additional studies are needed to extend the analysis to changes in the annual total N and S deposition, and to use

additional regional climate scenarios and model configurations to quantify the uncertainty in magnitude and spatial pattern of the deposition changes arising from uncertainties in simulated temperature and precipitation changes.

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Disclaimer

The views expressed here do not necessarily reflect those of the supporting or participating agencies, and no official endorsement should be inferred.

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