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Modeling atmospheric transport and fate of ammonia in North Carolina—Part II: Effect of ammonia emissions on fine particulate matter formation

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Abstract

Accurate estimates of ammonia (NH_3) emissions are needed for reliable predictions of fine particulate matter ($PM_{2.5}$) by air quality models (AQMs), but the current estimates contain large uncertainties in the temporal and spatial distributions of NH_3 emissions. In this study, the US EPA Community Multiscale Air Quality (CMAQ) modeling system is applied to study the contributions of the agriculture–livestock NH_3 (AL-NH_3) emissions to the concentration of $PM_{2.5}$ and the uncertainties in the total amount and the temporal variations of NH_3 emissions and their impact on the formation of $PM_{2.5}$ for August and December 2002.

The sensitivity simulation results show that AL-NH_3 emissions contribute significantly to the concentration of $PM_{2.5}$, NH_4^+, and NO_3^-; their contributions to the concentrations of SO_4^{2-} are relatively small. The impact of NH_3 emissions on $PM_{2.5}$ formation shows strong spatial and seasonal variations associated with the meteorological conditions and the ambient chemical conditions. Increases in NH_3 emissions in August 2002 resulted in $>10\%$ increases in the concentrations of NH_4^+ and NO_3^-; reductions in NH_3 emissions in December 2002 resulted in $>20\%$ decreases in their concentrations. The large changes in species concentrations occur downwind of the high NH_3 emissions where the ambient environment is NH_3-poor or neutral. The adjustments in NH_3 emissions improve appreciably the model predictions of NH_4^+ and NO_3^- both in August and December, but resulted in negligible improvements in $PM_{2.5}$ in August and a small improvement in December, indicating that other factors (e.g., inaccuracies in meteorological predictions, emissions of other primary species, aerosol treatments) might be responsible for model biases in $PM_{2.5}$.

Keywords: Ammonia emissions; Agriculture; Livestock; $PM_{2.5}$; CMAQ; Sensitivity study

1. Introduction

Ammonia (NH_3) is an important pollutant that plays a key role in several air pollution problems. It can create odors and have negative impacts on animal and human health. When deposited to ecosystems, NH_3 may cause over-enrichment of

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nitrogen, decrease in biological diversity, damage to sensitive vegetations, and acidification of soils (Fangmeier et al., 1994; Van der Eerden et al., 1998). As the most abundant gas-phase alkaline species in the atmosphere, NH_3 can neutralize sulfuric acid and nitric acid to form fine particulate matter with an aerodynamic diameter $\leqslant 2.5\,\mu m$ ($PM_{2.5}$), which is closely linked to health and climatic effects. In addition, NH_3 likely plays an increased role in $PM_{2.5}$ formation as the emissions of sulfur oxides and nitrogen oxides are reduced and a more stringent 24-h average $PM_{2.5}$ standard of $35\,\mu g\,m^{-3}$ is promulgated by the United States (US) Environmental Protection Agency (EPA) (Zhang et al., 2007).

Sulfate (SO_4^{2-}) and nitrate (NO_3^-) aerosols are two major inorganic components of $PM_{2.5}$ in the eastern US (EPA, 1996). A recent study shows that for the eastern US, a reduction in sulfate dioxide (SO_2) may not be as effective as it is often assumed in reducing PM mass, as a reduction in SO_4^{2-} concentrations results in more free NH_3 available for reaction with nitric acid (HNO_3) to produce ammonium nitrate (NH_4NO_3) particles (West et al., 1999). The accuracy of NH_3 emissions can have a large effect on air quality model (AQM) predictions of aerosol SO_4^{2-}, NO_3^-, and ammonium (NH_4^+) concentrations (Mathur and Dennis, 2003). However, large uncertainties exist in NH_3 emission inventories in both total annual emissions and the monthly, daily, and diurnal variations, since NH_3 emissions are largely from non-point sources such as livestock operations and fertilized fields, all those sources are difficult to be directly measured (Pinder et al., 2006). Current seasonally varied NH_3 emission inventories have been developed using several advanced methods including inverse methods (e.g., Gilliland et al., 2003), process-base models (e.g., Pinder et al., 2004a, b), and hybrid approaches (e.g., Skjøth et al., 2004).

Major emission sources of NH_3 include animal and human wastes, synthetic fertilizers, biomass burning, and soil biogenic emissions (Bouwman et al., 1997) . North Carolina (NC) is one of the largest agricultural product states in the US, ranking the 2nd in hogs, 2nd in turkeys, and 5th in broilers. NH_3 emissions from hog farms account for more than 80% of total NH_3 emissions in NC (Wu et al., 2007). Most hog farms are located in the coastal plain region of the state or the southeast corner covering Bladen, Duplin, Greene, Lenoir, Sampson, and Wayne counties.

In this study, the atmospheric transport and fate of NH_3 are studied using a three-dimensional (3-D) transport and chemistry model. Part I of our studies (Wu et al., 2007) describes the model configurations, evaluation protocols and databases used, and the operational evaluation for meteorological and chemical predictions. In Part II, we describe the sensitivity simulations under various emission scenarios. Our objectives are to quantify the contribution of NH_3 emissions to the formation of $PM_{2.5}$ and its composition and assess the uncertainties in the total amount and temporal variations of NH_3 emissions and their impact on $PM_{2.5}$ predictions.

2. NH_3 emission inventories and sensitivity simulation design

2.1. Baseline NH_3 emission inventories

The baseline simulations at a 4-km grid spacing are conducted for August and December 2002 using the 5th Generation Penn State/NCAR Mesoscale Model (MM5) version 3.7, the Carolina Environmental Program's (CEP) sparse matrix operation emission (SMOKE) modeling system version 2.1, and the US EPA Models-3 Community multiscale air quality (CMAQ) modeling system version 4.4. Detailed configurations can be found in Wu et al. (2007). The baseline 4-km emissions are generated based on the NH_3 emission inventory developed under the Visibility Improvement State and Tribal Association of the Southeast (VISTAS) program (http://www.vista-sesarm.org.asp) (referred to as NH_3-VISTAS hereafter). The Carnegie Mellon University (CMU) NH_3 model version 3.6 is used to calculate NH_3 emissions in NH_3-VISTAS that have been improved from previous emission estimates based on the EPA 1999 National Emission Inventories version 2 with activity and growth data of CMU NH_3 model version 3.1 (Abraczinskas, 2005). NH_3-VISTAS uses the United State Department of Agriculture (USDA) 2002 census county-level livestock amounts and process-level distribution for dairy cattle, beef cattle, swine, goats, poultry, and turkeys for livestock activity levels, and the 2002 fertilizer application activity data of the Association of American Plant Food Control Officials. Other NH_3 sources (e.g., waste treatments, motor vehicles, etc.) are described in CMU model by Strader et al. (2005). NH_3-VISTAS includes all NH_3 sources except the domestic animal emissions