Inverse Modeling to Estimate Seasonal Ammonia Emissions

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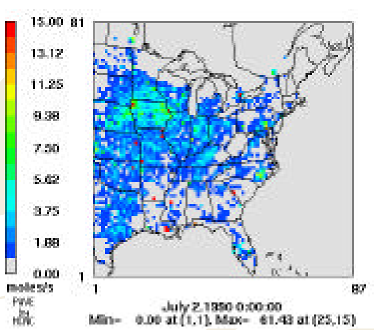
Introduction

Ammonia (NH_3) emissions are a vital input for modeling regional patterns of nutrient deposition, visibility, fine particulates, and acid precipitation. Approximately 85% of NH_3 emissions come from agricultural sources, which are thought to have a strong seasonal signal. Yet, most current inventories assume no seasonal variability and the annual emission estimates are suspect. Within a regional modeling framework, we assess the accuracy of NH_3 emissions and their seasonal variability. For each season, a Kalman filter inverse modeling technique[1,2,3] is used to deduce NH_3 emissions that will result in modeled NH_4^+ wet concentrations that agree optimally with observed concentrations.

Annual U.S. Ammonia Emissions (1997)	
	million kg NH_3
Livestock:	
Cattle[4]	2,267
Swine[4]	563
Poultry[4]	343
Other animals	72
Fertilizer application[5]	615
Industrial/other[5]	611
TOTAL	4,471

Figure 1. Monthly average NH_3 emission estimates used in this study.



Approach

We compare model and observed wet concentrations of ammonium (NH_4^+) using a Kalman filter inverse modeling approach.[1,2,3] This method is applied to deduce ammonia emissions that should provide modeled NH_4^+ wet concentrations that optimally agree with observations. The model, observations, and inverse modeling technique are briefly described below.

Air Quality Model:

The USEPA Models-3 Community Multiscale Air Quality Model (CMAQ)[6] is a multi-pollutant air quality model that includes ozone and particulate matter (or aerosols). CMAQ is configured for this inverse modeling study as follows:

- Eastern US domain (as shown in Figure 1)
- 36 km horizontal grid resolution, 21 vertical layers
- June 5 – July 3, 1990 completed
- January 9 – February 7, 1990 partially completed

Observations:

We use weekly NH_4^+ wet concentration samples collected by the National Acid Deposition Program (NADP) Network. NADP scientists have observed an average low bias of ~15% in the weekly collected samples of $[NH_4^+]$,[7] which we have incorporated into the observed data for this study. More information about this network of data can be obtained from http://nadp.sws.uiuc.edu.

Inverse Modeling Method:[1,2,3]

m = # of source regions

$$\Delta E(m) = G_t(m \times n)\Delta \boldsymbol{c}_t(n) \qquad \text{(Equation 1)}$$

n = # of obs

where : $\Delta E = E_{t,i+1} - E_{t,i}$

$$\Delta \boldsymbol{c}_t = \boldsymbol{c}_t^{obs} - \boldsymbol{c}_{t,i}^{model}$$

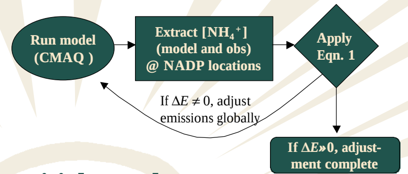
$$G_t = C_{t,i}P_t^T\left(P_tC_{t,i}P_t^T + N_t\right)^{-1}$$

$$C_{t,i+1} = C_{t,i} - G_{t,i}P_tC_{t,i}$$

$$P_t = \frac{\partial \boldsymbol{c}_t}{\partial E_t}$$

N_t : variance in measurement error

In this approach, the following steps are performed:

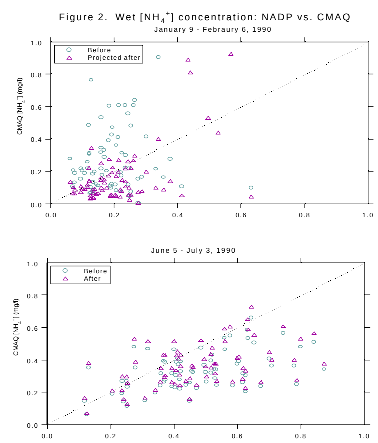


Initial Results

Initial results are available for June 1990 and partially for January 1990. They suggest NH_3 emissions should be adjusted as follows:

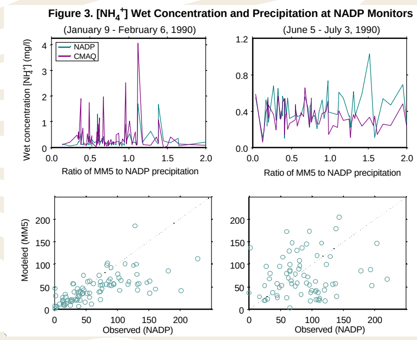
- **(0.42 * annual NH_3 emissions) for January 1990**
- **(1.11 * annual NH_3 emissions) for June 1990**

For June 1990, $[NH_4^+]$ before and after emission adjustments are shown in Figure 2. Since the simulation is currently being completed, the January 1990 results only show the before emission adjustment results and the projected results using the filter adjustment. Ultimately, we will develop emission estimates for each season of 1990.



Issues

Non-emission related errors in the air quality model can influence the results obtained with the Kalman filter. An important issue is the model's ability to simulate precipitation. In Figure 3, modeled and observed $[NH_4^+]$ is sorted for June 1990 by the ratio of modeled/observed precipitation.



In June, CMAQ predictions of $[NH_4^+]$ have more errors when the modeled precipitation is too high versus NADP observations. January 1990 results should be less affected by this problem. Overpredictions are evident for January, 1990; however, they are not associated with precipitation biases according to Figure 3. Additionally, model precipitation agrees better with NADP observed values for January than June 1990 (Figure 3). We are currently studying new statistical techniques that could better account for model uncertainties such as these.

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