# EVALUATING AMMONIA ($NH_3$) PREDICTIONS IN THE NOAA NATIONAL AIR QUALITY FORECAST CAPABILITY (NAQFC) USING GROUND-BASED AND SATELLITE-BASED MEASUREMENTS ON A NATIONAL SCALE

William Battye,* Casey Bray, and Viney Aneja
Department of Marine, Earth, and Atmospheric Sciences North Carolina State University, Raleigh, NC

Daniel Tong, Pius Lee, and YouHua Tang
NOAA Air Resources Laboratory, College Park, Maryland, MD

## 1. INTRODUCTION

Ammonia ($NH_3$) gas in the atmosphere contributes to the formation of airborne fine particulate matter ($PM_{2.5}$), which is associated with adverse health effects. Atmospheric $NH_3$ also deposits into terrestrial and aquatic ecosystems, potentially contributing to eutrophication and impacts on species diversity. The U.S. National Oceanic and Atmospheric Administration (NOAA) is responsible for forecasting elevated levels of $PM_{2.5}$ within the framework of National Air Quality Forecast Capability (NAQFC), and these forecasts require reliable estimates of precursor $NH_3$ concentrations. The Community Multiscale Air Quality (CMAQ) model is used to simulate emissions of $NH_3$, atmospheric transport, and conversion of $NH_3$ to $PM_{2.5}$. CMAQ is also used to calculate the deposition of $NH_3$ and other nitrogen compounds to sensitive ecosystems. However, emission, transport, and deposition processes for $NH_3$ are subject to considerable uncertainty.

The objective of the current research is to design a framework for using satellite-based measurements improve CMAQ predictions of $NH_3$. We also use ground-based measurements to test the validity of satellite observations and to evaluate model performance. Ultimately, we expect refined predictions of $NH_3$ to result in improved predictions of $PM_{2.5}$ and nitrogen deposition.

## 2. BACKGROUND

Agricultural sources account for about 90% of atmospheric $NH_3$ emissions in the U.S. Emissions emanate primarily from animal waste management and synthetic nitrogen fertilizer application. Other sources include chemical and petroleum processes, wastewater treatment, forest fires, and the use of $NH_3$ in control systems for reducing $NO_X$ emissions such as selective catalytic reduction (SCR). Emissions estimates for $NH_3$ are subject to considerable uncertainty, and prior model-to-monitor comparisons suggest emissions may be underestimated in some regions of the U.S.

## 3. METHODS AND DATA

### *3.1 Modeling*

CMAQ version 5.0.2 was used for air quality modeling. Meteorological inputs are derived from the Nonhydrostatic Multiscale Model on B-grid (NMMB). The modeling domain is the continental U.S. and the time period simulated was July 2011. Model grid horizontal resolution is 12 km, and 42 vertical layers are used, starting with a lowest layer 8 m above ground level. The vertical domain top is 50 hPa. The AERO5 model is used for aerosol chemistry, and deposition is based on the M3Dry module.

$NH_3$ emissions are derived from the U.S. Environmental Protection Agency (EPA) 2011 National Emissions Inventory (NEI). The NEI estimates emissions at the county level based on daily meteorological conditions. County-level emissions are allocated to the CMAQ grid using the Sparse Matrix Kernel Emissions (SMOKE). SMOKE diurnal allocation factors were also used to generate hourly emissions estimates. The NAQFC CMAQ modeling framework does not yet incorporate bi-directional flux of $NH_3$ from crops treated with chemical fertilizer.

### *3.2 Ground-level Measurements*

The Ammonia Monitoring Network (AMoN) measures concentrations of atmospheric $NH_3$ on a 2-week average basis using passive diffusion samplers. We obtained data from 48 AMoN

*Corresponding author: William Battye, North Carolina State University, Raleigh, NC; and EC/R Division of SC&A Incorporated, Chapel Hill, NC 27514; e-mail: whbattye@ncsu.edu

sampling locations active in July 2011. We also obtained data on the mass of ammonium ions ($NH_4^+$) in the particle phase chemically-speciated $PM_{2.5}$ monitors located near the AMoN sites. The Clean Air Status and Trends Network (CASTNet) includes 27 sites located within 12 km of AMoN sites active in 2012. The Interagency Monitoring network for Protected Visual Environments (IMPROVE) includes another 20 monitors within 12 km of AMoN sites.

### *3.3 Satellite-Derived $NH_3$ Measurements*

We obtained daytime measurements of atmospheric $NH_3$ during July 2011 from two satellite platforms. The Tropospheric Emission Spectrometer (TES) on the Aura satellite makes measurements over the continental U.S. between 1:00 and 3:00 pm local time. The Atmospheric Science Data Center at the NASA Langley Research Center publishes TES retrievals of the total atmospheric column loading of $NH_3$ and the vertical profile of atmospheric $NH_3$ concentrations (Shepherd *et al,* 2011). These retrievals also include estimates of the degrees of freedom for signal (DOFS). We use TES retrievals with DOFS greater than or equal to 0.5, or a thermal contrast of greater than or equal to 7 K.

The Infrared Atmospheric Sounding Interferometer (IASI) on the MetOp satellite makes measurements over the continental U.S. between 9:40 and 10:20 am local time. The Atmospheric Spectroscopy group of the Université Libre de Bruxelles, Belgium has retrieved total atmospheric column loadings of $NH_3$ from IASI measurements (Whitburn *et al*, 2015). These retrievals also include estimates of the measurement error. We use IASI retrievals where the relative error is less than 100% or the absolute error is less than 1.4 mg/m$^2$ ($5\times10^{-15}$ molecules/cm$^2$)

The IASI instrument scans a broader angle than the TES instrument, and takes more measurements in a given satellite pass. For the month of July 2011, 209 TES retrievals were available meeting the screening criteria, while 58,341 IASI retrievals were available.

For each satellite retrieval, we compute a corresponding model value for $NH_3$ by interpolating from the four surrounding model grids and the two nearest time steps.

### *3.4 Model-to-Monitor Comparisons*

Normalized mean bias (NMB) of model predictions is computed as follows:

$$NMB = \frac{1}{N}\frac{\sum_{i=1}^{N}[C_{mod}(i) - C_{obs}(i)]}{\sum_{i=1}^{N} C_{obs}(i)}$$

where $C_{mod}(i)$ and $C_{obs}(i)$ are, respectively, the model prediction and the observed concentration at a given location and time, and N is the number of observations.

## 4. RESULTS AND DISCUSSION

Figure 1 plots ground level measurements of $NH_3$ from the AMoN network against model predictions. Figure 2 plots ground level measurements of particulate $NH_4$+ from the CASTNet and IMPROVE networks against model predictions. Figures 3 and 4, respectively, plot TES and IASI retrievals of total column $NH_3$ against model predictions.

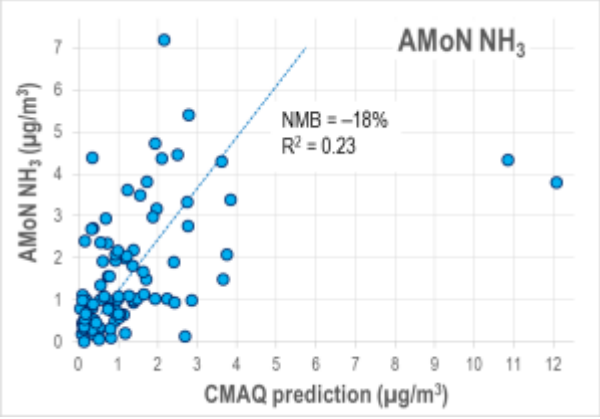


Figure 1. Ground level $NH_3$ measurements (AMoN) compared with model predictions.

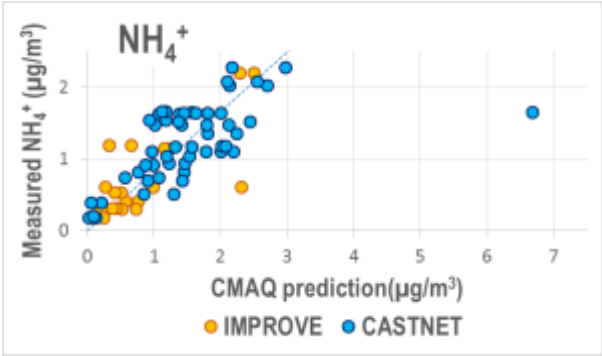


Figure 2. $NH_4^+$ measurements (IMPROVE and CASTNET) vs. model predictions.

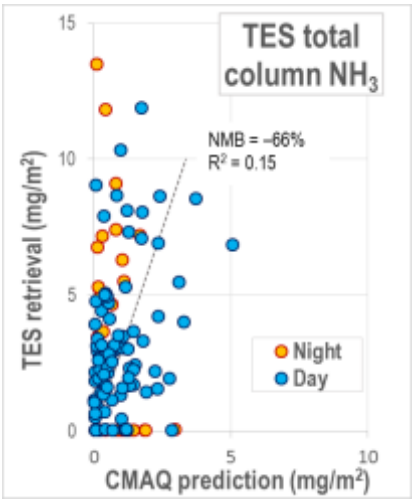


Figure 3. TES satellite retrievals of total column $NH_3$ plotted against model predictions.

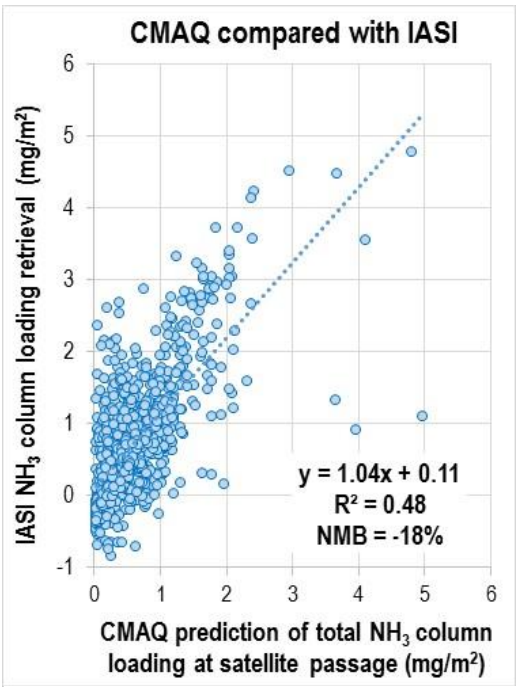


Figure 4. IASI retrievals of total column $NH_3$ plotted against model predictions. (Each point represents an average over a 120 km x 120 km region.)

All of the measurement data sets are subject to significant variability, and there is low correlation between model predictions and particular corresponding measurements. Model evaluation results also differ somewhat for the measurement data sets. Comparisons with AMoN and IASI show an average negative bias of about 18% (underprediction). Comparisons with TES indicate a larger negative average bias. Some of this bias may stem from errors in predicting short-term concentrations at the time of satellite passage.

Model prediction bias is subject to spatial variation at large and small scales. Figure 5 plots spatial variations in model bias as indicated by AMoN and TES comparisons. The figure maps the average difference between measured and modeled concentrations at the surface for all observations in July 2011. (As noted above, TES retrievals estimate the vertical profile of $NH_3$, including the concentration at the surface). The model-measurement differences are superimposed against a background layer showing the modeled concentration.

Figure 6 maps spatial variations in the average difference between measured and modeled total column concentrations of $NH_3$. In order to analyze regional patterns, IASI observations were averaged to a spatial resolution of 120 km x 120 km (10 x 10 CMAQ modeling grids). Figure 6 plots the spatially averaged model-measurement differences, superimposed against a background layer showing the modeled column loading.

As Figures 5 and 6 show, results of the regional comparisons vary among the three measurement datasets. However, all three datasets suggest that the model may be underpredicting atmospheric $NH_3$ in the Midwest and in the Central Valley of California, and overpredicting $NH_3$ in parts of the Southeast U.S.

Differences between modeled and measured concentrations may stem from uncertainties in the $NH_3$ emissions inventory, spatial and temporal allocation methods used in the inventory, model deposition processes, or other model processes. A recently developed bidirectional flux algorithm for $NH_3$ could be expected to raise $NH_3$ concentrations in the summer months, thereby ameliorating some underprediction. However, it must also be noted that model bias estimates are subject to considerable uncertainty because of large variabilities associated with all of the measurement data sets.

## 5. ACKNOWLEDGEMENTS

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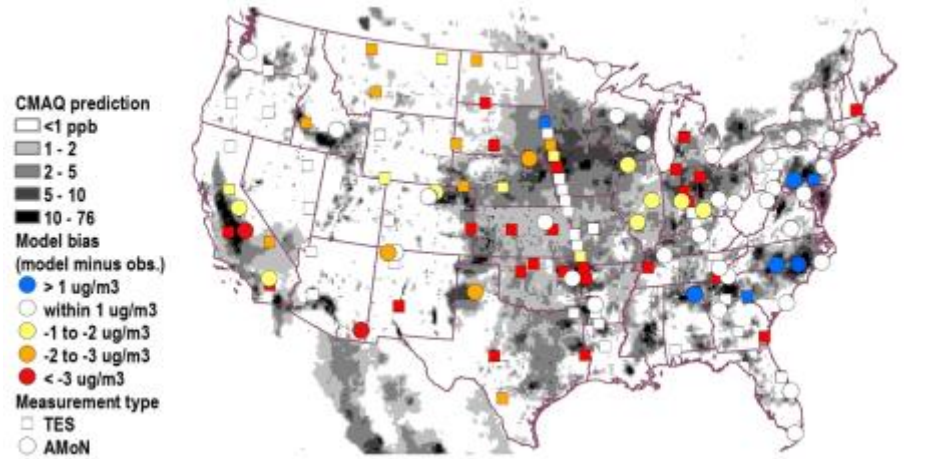


Figure 5. Spatial variation in model prediction error based on AMoN measurements and TES retrievals of surface concentrations over a background of model predictions.

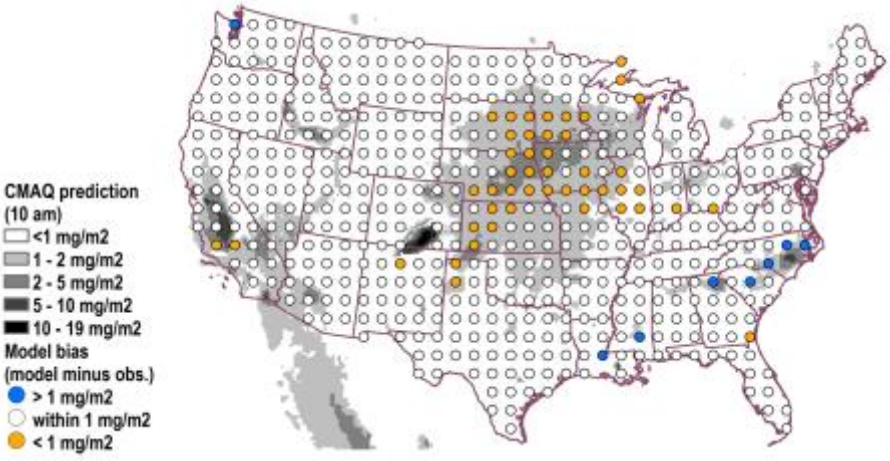


Figure 6. Spatial variation in model prediction error based on IASI retrievals of total column concentration over a background of model predictions. Each icon represents at least 31 observations, and an average of 108 observations.

retrievals, and advice on filtering and analyzing IASI data. We also acknowledge the NOAA National Air Quality Forecast Program, and the North Carolina State University Air Quality Research Group.

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