Diagnostic Evaluation of Emissions via Top-down Inverse Modeling

Robert Pinder, Sergey Napelenok, and Alice Gilliland

2009 AMAD Peer Review: Session 2
Model Evaluation: Establishing Model's Credibility

Environmental Issue

Emission uncertainties have direct impacts on model evaluation results, but they also affect regulatory applications of air quality models. For example, ammonia (NH_3) emission biases could impact model-estimated $PM_{2.5}$ decreases associated with SO_2 emission reductions (Pinder et al., 2008; Dennis et al., 2008). Mobile NO_x emissions biases can cause models to underestimate O_3 changes from utility sector decreases in NO_x emission (Gilliland et al., 2008).

- ♦ Emissions are among the largest uncertainties for air quality modeling.
- ♦ Except for directly measured NO_x, SO_2, and CO_2 emissions from utilities, emissions cannot be directly measured across regional domains.
- ♦ Many emission sources are diffuse and difficult to estimate.

In situations where emission uncertainties are large, impacts on predicted air pollutant concentrations can be substantial.

To complement the tremendous efforts that go into bottom-up emission inventories, "top-down" methods are needed to evaluate emissions using relevant ambient concentrations.

Research Objectives

As part of the Division's diagnostic evaluation effort, inverse modeling has been used to evaluate and refine emission estimates. There have been two main study areas for inverse modeling:

- ♦ NH_3 emissions, which have large uncertainties both in seasonal distribution and total budget (Gilliland et al., 2003, 2006; Pinder et al., 2006).
- ♦ NO_x emissions using satellite data and CMAQ-DDM (Napelenok et al., 2008a; 2008b; Poster 2.3).

General research objectives for these inverse studies:
(1) Develop inverse methods
(2) Assess emissions for bias
(3) Do inverse-derived emission changes improve model results?
(4) Inform emission inventory updates

Modeling Approach

The following are used in inverse modeling applications to evaluate or infer air quality emissions:

- Chemical transport model
- Estimated change of pollutant concentrations with emission change – discussed more below
- Uncertainty estimates for emissions and concentrations

This inverse modeling method is based on an adaptive-iterative version of the Kalman filter (Haas-Laursen et al., 1996; Gilliland and Abbitt, 2001):

$$E_t^{posterior}(m) - E_t^{prior}(m) = G_t(m \times n)\left(\chi_t^{obs} - \chi_t^{mod}\right)$$
$$G_t = S_a K^T \left(K_t S_a K_t^T - S_\Sigma\right)^{-1} \quad (1)$$

E = emissions for each source region(s); posterior refers to emission estimate from applying equation (1); prior refers to previous "best guess" emissions
m = # of source regions; n = # of observations
S_a = variance of error in emissions
S_Σ = variance of error in concentrations (observations and model estimates)

The Jacobian K_t is a model-estimated sensitivity of the pollutant of interest to a primary or precursor emission:

$$K_t = \frac{\partial\left(\chi_t^{mod} - \chi_{t-1}^{mod}\right)}{\partial t} = \frac{\partial\left(\chi_t^{mod}\right)}{\partial t}\ (*) \quad (2)$$

(*) Equation 2 can only be reduced if either (a) the modeled concentration at t-1 is equal to observations, or (b) the model concentration at time t is independent of t-1 (i.e., initial conditions), as demonstrated by Gilliland and Abbitt (2001).

K_t is often estimated via "brute force" comparison of model simulations with a small emission change. The NH_3 study presented here used this approach. A new approach, introduced here, uses the CMAQ-DDM (Poster 2.3) to quantify K_t. Either way, it is critical to establish the relationship between the emissions and observed chemical quantities, such as deposition or ambient concentration.

The following steps (Figure 1) are followed in these studies:
(1) Quantify K_t
(2) Apply Equation 1 for "prior" case model simulation
(3) Re-run model simulation with posterior adjusted emissions
(4) Re-apply Equation 1, and repeat until $\Delta E \approx 0$.

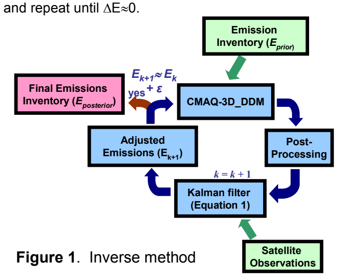

Figure 1. Inverse method

Results and Discussion

Agricultural NH_3 Emissions

Agricultural emissions are the largest NH_3 source, yet the seasonal variation of these emissions is undetermined. Gilliland et al. (2006) developed top-down estimates of seasonal factors for NH_3 emissions using the inverse method described here, the CMAQ model, NADP NH_4^+ wet concentration data, and the most advanced emission inventories as a priori estimates (Figure 2), including seasonal factors developed by Goebes et al. (2003) for fertilizer, Pinder et al. (2004) for dairy cattle, and Gilliland et al. (2003) for all other sources.

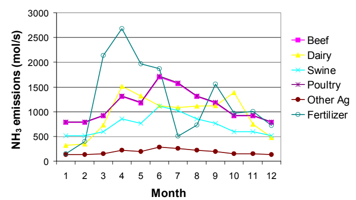

Figure 2. NH_3 emissions from agriculture

Errors in the seasonal variability of NH_3 emissions cause large errors in simulated NH_4^+ aerosol and $PM_{2.5}$ nitrate predictions. The inverse-derived emission estimates improved CMAQ-predicted NH_4^+ wet deposition as well as the $PM_{2.5}$ nitrate model error (Figure 3) – an independent check on the quality of the updated emissions. Further analysis concluded that improvements in top-down assessments are limited until ambient NH_x (NH_3 + NH_4^+) are available.

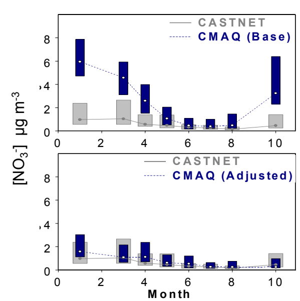

Figure 3. CMAQ ambient $PM_{2.5}$ nitrate and CASTNET observations before (top) and after (bottom) top-down estimates of NH_3 emissions (Gilliland et al., 2003).

Urban NO_x Emissions

In the NH_3 study, the entire Eastern U.S. was treated as a single source region. Recently, an adapted approach was tested where separate urban and rural source regions for NO_x were tracked using CMAQ-DDM. This provided the K_t sensitivity to emission changes by source region (Equation 2). The NO_2 column retrievals from SCIAMACHY provide the denser data needed to resolve the source regions.

Key results from Napelenok et al. (2008):

- Vehicle NO_x emissions were too high in urban Birmingham and Atlanta
- Independent ambient data confirm model improvement (Figure 4).
- NO_x biases in free troposphere impact comparisons of NO_2 columns in rural areas (Figure 5).

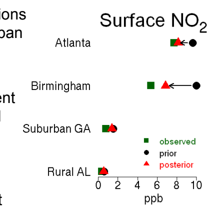

Figure 4. Comparison of surface NO_2 at four SEARCH sites.

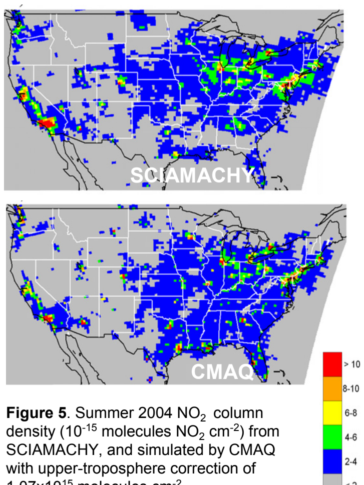

Figure 5. Summer 2004 NO_2 column density (10^{-15} molecules NO_2 cm^{-2}) from SCIAMACHY, and simulated by CMAQ with upper-troposphere correction of 1.07×10^{15} molecules cm^{-2}.

Conclusions

- Emission uncertainties substantially influence air quality model performance.
- Inverse modeling studies have effectively demonstrated how "top-down" methods can be used for regional-scale air quality emission assessments.
- Seasonal NH_3 emissions estimates can substantially impact inorganic $PM_{2.5}$
- Further advances in inverse modeling can be made using satellite data and instrumented models
- Continued evaluation of mobile NO_x emissions is needed in urban core areas.

Future Directions

- As NH_x (NH_3+NH_4^+) data become available, new inverse assessments of NH_3 with detailed source regions is warranted.
- With more highly resolved satellite data and improved model chemistry, additional NO_x inverse studies may be conducted.
- Elemental carbon and carbon monoxide are additional candidates for inverse studies because of the high level of uncertainty and the low impact of chemical processes.

Impact

Top-down emission evaluation provides important feedback to emission inventory development. NH_3 seasonal estimates have been incorporated directly into the SMOKE emissions system. The development of these inverse methods has encouraged the implementation of NH_x monitoring networks. NO_x inverse modeling can improve estimates of urban vehicle emissions, the largest NO_x emission source.

Collaborators

Randall Martin, Dalhousie University