Resolving NO_x Emission Inventory Biases Using Discrete Kalman Filter Inversion, Direct Sensitivities, and Satellite-based NO_2 Columns

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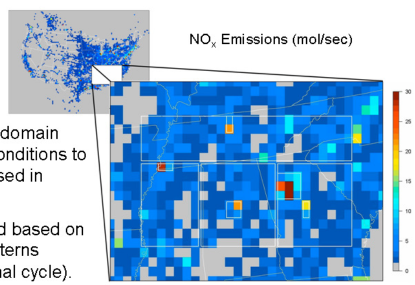
Abstract

Column NO_2 concentrations observed by the SCIAMACHY satellite in the southeastern United States during the summer months of 2004 were matched with the results of the CMAQ model. Assuming that the major discrepancies between NO_2 observations and model predictions were due to the uncertainties in the NO_x emission inventory used to drive the air quality model, discrete Kalman filter inversion was performed to arrive at possible emission biases across several predefined geographic regions. The relationship between NO_x emissions and NO_2 concentrations was represented with sensitivities computed using the Decoupled Direct Method in 3D.

The majority of work in inverse modeling has involved global scale models and/or inert species. Here, a method is presented that can be used as an additional tool for examining the quality of emission inventories for regional-scale air quality modeling applications.

Modeling Domain and Source Regions



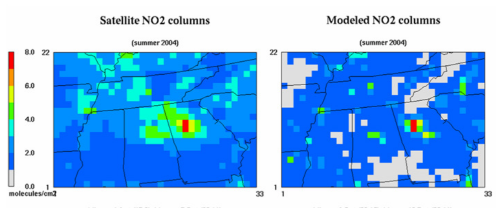
The continental U.S. domain provided boundary conditions to the nested domain used in detailed analysis.

Regions were defined based on similar emissions patterns (magnitude and diurnal cycle).

Observed and Modeled NO_2 Columns

Summertime averages during the time of the satellite overpass (1600) were computed taking into account only the geographical extend of the coverage during each day between June 1, 2004 and August 31, 2004.

The comparison revealed lower NO_2 model predictions in rural areas with low emissions and higher model predictions in the urban centers.

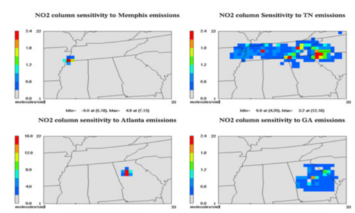


Decoupled Direct Method in 3D

DDM 3-D computes first-order, semi-normalized sensitivities to perturbations in an input field (here, regional emissions of NO_x):

$$S_{NO_2,NO_{x,r}}(\bar{x},t) = \frac{\partial C_{NO_2}(\bar{x},t)}{\partial p_{NO_{x,r}}}$$

DDM 3-D provides gridded sensitivity fields simultaneously with concentrations as the model progresses in time.



CMAQ implementation is discussed in Cohan et al., 2005 and Napelenok et al., 2006.

Discrete Kalman Filter

The Kalman Filter is an optimization technique used to estimate discrete time series and states that are governed by sets of linear differential equations. The linearity assumption is overcome by applying the technique iteratively.

The emissions vector evolves as follows:

$$\bar{E}_{t,k+1} = \bar{E}_{t,k} + \mathbf{G}_{t,k}(\bar{\chi}_t^{obs} - \bar{\chi}_t^{mod})$$

Gain matrix is computed from:

$$\mathbf{G}_{t,k} = \mathbf{C}_{t,k}\mathbf{P}_t^T(\mathbf{P}_t\mathbf{C}_{t,k}\mathbf{P}_t^T + \mathbf{N}_t)^{-1}$$

k	= iteration
$\bar{E}_{t,k}$	= Emissions vector
$\mathbf{G}_{t,k}$	= Gain matrix
$\bar{\chi}_t^{obs}$	= Observations vector
$\bar{\chi}_t^{mod}$	= Modeled value vector
$\mathbf{C}_{t,k}$	= Covariance of error of emissions
$\mathbf{P}_t$	= Normalized sensitivity
$\mathbf{N}_t$	= Noise matrix

The covariance of error matrix is updated after each iteration:

$$\mathbf{C}_{t,k+1} = \mathbf{C}_{t,k} - \mathbf{G}_{t,k}\mathbf{P}_t\mathbf{C}_{t,k}$$

More information can be found in Gilliland and Abbitt, 2001 and Rodgers, 2000.

References

Cohan, D.S., Hakami, A., Hu, Y., Russell, A.G., 2005. Nonlinear response of ozone to emissions: source apportionment and sensitivity analysis. *Environmental Science & Technology* 39, 6739-6748.

Gilliland, A., Abbit, P.J., 2001. A sensitivity study of the discrete Kalman filter (DKF) to initial condition discrepancies. *Journal of Geophysical Research* 106, 17939-17952.

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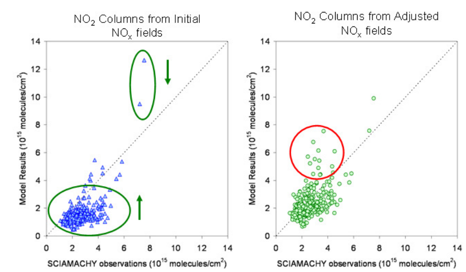
Results

The method converges on a solution with only a few iterations. In general, rural emissions are increased and urban emissions are decreased.

Region	NO_x Emissions[1] (mol/sec)	Adjusted NO_x Emissions (mol/sec)
Urban Centers		
Memphis, TN	36.5	33.2
Nashville, TN	28.8	27.7
Birmingham, AL	35.6	30.5
Atlanta, GA	155.3	131.3
Macon, GA	25.3	13.1
Surrounding Areas		
Tennessee	369.2	663.9
Mississippi	152.8	222.7
Alabama	181.9	392.0
Georgia	158.5	342.5

[1]Average hourly rate during the time of satellite overpass in only the 1st layer of the model.



Discussion

This method was able to efficiently adjust regional emissions of NO_x in order to closer match CMAQ modeled NO_2 levels with those observed by the SCIAMACHY satellite in southeastern United States during the summer of 2004.

Concentrations in parts of some regions were pushed much higher than satellite observations. This occurred near cells with emissions significantly higher than region averages. For example, in the area near Chattanooga, TN, emissions were increased with the rest of the Tennessee region. This problem can be avoided by introducing more finely defined source regions.

Discrepancies in the modeled and observed NO_2 concentrations are not due solely to estimations of emissions at the ground level. Uncertainties in the chemical processes, emissions aloft from lightning sources and airplanes, and meteorological predictions also contribute, but were assumed to be negligible.

Furthermore, while it is possible to perform a similar analysis for shorter time averages (days), instead of the entire summer, inconsistent satellite coverage makes it difficult to constrain the optimization equations. The more recently available OMI satellite instrument has daily coverage and will help alleviate this problem.

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