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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A method for evaluating spatially-resolved NO_x emissions using Kalman filter inversion, direct sensitivities, and space-based NO_2 observations" *by* S. L. Napelenok et al.

Anonymous Referee #1

Received and published: 14 May 2008

This paper is within the scope of ACP. It presents an approach to determining NOx emissions in the southeastern United States using SCIAMACHY satellite observations of NO2 vertical columns, the CMAQ regional chemical-transport model, and the Kalman filter inversion technique. This work is a relatively new application of this methodology to the estimation of regional and urban NOx emissions. The authors present some interesting test cases using pseudo-observations and demonstrate some of the sensitivities to the inversion parameters. Their inversion suggests that 2004 NOx emissions in southeastern US urban areas were about 10% lower than the EPA 2001

bottom-up inventory estimates. They also find a significant model underestimate of upper tropospheric NOx, which they attribute to lightning sources.

I find the paper good in general. Reviewer #2 has already raised a number of important concerns with the analysis and methodology. Below I list only my additional comments beyond those brought up by that Reviewer. I recommend this paper for publication in ACP if all concerns expressed below and by Reviewer #2 are addressed.

Specific comments:

P6471, L2-5: Regional air quality models require well-quantified emissions for other purposes besides the two specific applications mentioned. Two more uses of these models that readily come to mind are assessing of our knowledge of atmospheric processes (such as chemical mechanisms, transport schemes, boundary layer dynamics, etc) and real-time forecasting of air quality. Please make this list a bit more inclusive.

P6471-2: Although not an *inverse* modeling study, Kim et al. 2006 used SCIAMACHY and GOME data along with the WRF-Chem model in a top-down approach to evaluate another EPA NOx emission inventory and examine trends in US NOx emissions on a regional scale. A full citation is included below and should be referenced. Kim, S.-W., Heckel, A., McKeen, S. A., Frost, G. J., Hsie, E.-Y., Trainer, M. K., Richter, A., Burrows, J. P., Peckham, S. E., and Grell, G. A.: Satellite-observed US power plant NOx emission reductions and their impact on air quality, Geophys. Res. Lett., 33, L22812, doi:10.1029/2006GL027749, 2006.

P6473, L1-6: Even after reading Gilliland et al 2008, it was not clear to me whether the monthly, or at least seasonally, appropriate power plant NOx emissions were used. Adjusting power plant NOx emissions to account for reductions between 2001 (the basic NEI inventory) and 2004 (the year of CEMs data used) using only annual average values would still overestimate NOx emissions from this sector, since additional NOx controls are used only in the summer at southeastern US power plants. This could contribute to the downward adjustment needed in the NOx inventory across many of

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the source regions. Please clarify this point.

P6481, L4: Prior to Hudman et al. 2007, Cooper et al. 2006 published a report of the extensive NOx production from lightning over North America during the summer of 2004. Please include the following citation. Cooper, O. R., et al.: Large upper tropospheric ozone enhancements above midlatitude North America during summer: In situ evidence from the IONS and MOZAIC ozone measurement network, J. Geophys. Res., 111, D24S05, doi:10.1029/2006JD007306, 2006.

P6481-6484: A spatially uniform increase in NO2 columns was used to account for missing lightning NOx production in the model. Using lightning flash measurement networks, could the authors get a better sense of the spatial distribution of lightning (and therefore of this additional NOx source)? Even if such an analysis is beyond the scope of the paper, please comment on how a spatially inhomogeneous upper tropospheric NOx source might be included in your analysis.

P6484, L6-8: The adjustments to the inventory in the Mississippi source region are also outside the specified uncertainty of the inventory. Only the Macon region is specifically mentioned here.

Technical corrections:

P6470, L8-10: suggest rewrite as follows: *as constrained by observations of NO2 column densities derived from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) satellite instrument.*

P6470, L21: add *a* before *combination*

P6470, L24: change *from* to *by*

P6471, L10: should read *activity-specific emission factors*

P6475, Eq. 5: Is the factor of 0.1 in the off-diagonal elements of the initial covariance matrix an arbitrary value? Perhaps mention the reason for this factor here.

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P6476, L6: add *of* after *nature*

P6476, L9: add *the* after *include*

P6477, L20: Note the error factor here too; currently it appears only in the caption to Fig. 3.

P6479, L13: Add *the* before *opportunity*

P6482, L7: Add *the* before *inverse*

P6484, L6: Omit *,* after *factors*

Fig.1: It might be useful to identify the source regions on the map itself for the benefit of those not well-versed in US geography.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 6469, 2008.