Atmos. Chem. Phys. Discuss., 8, S3996–S4001, 2008
www.atmos-chem-phys-discuss.net/8/S3996/2008/

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.

S. L. Napelenok et al.

Received and published: 23 June 2008

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.

To follow the reviewer's suggestion, this text was changed as follows:

Regional air quality modeling has been used to develop control strategies

designed to reduce levels of pollutants such as ozone and particulate matter. Models have been used to assess our knowledge of atmospheric processes, including chemical and physical transformations of air pollutants, and to forecast air quality. More recently, results of regional models have been integrated into epidemiological studies that aim to assess the health impacts of atmospheric pollutants (Knowlton et al., 2004). All of these applications rely on well quantified emission inputs.

*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 NO_x emission inventory and examine trends in US NO_x 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.*

The citation was added to the text as follows:

These data, as well as ground-based and other observations, have been used previously in inverse modeling of "top-down"; inventories, but typically on the global scale (Martin et al., 2003; Müller and Stavrakou, 2005), and less frequently on the regional scale (Blond et al., 2007; Kim et al., 2006; Quélo et al., 2005; Konovalov et al., 2006; Konovalov et al., 2008; Wang et al., 2007).

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 NO_x emissions were used. Adjusting power plant NO_x emissions to account for reductions between 2001 (the

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basic NEI inventory) and 2004 (the year of CEMs data used) using only annual average values would still overestimate NO_x emissions from this sector, since additional NO_x controls are used only in the summer at southeastern US power plants. This could contribute to the downward adjustment needed in the NO_x inventory across many of the source regions. Please clarify this point.

In this work, as well as in Gilliland et al., 2008, daily power plant NO_x emissions collected from CEMs were used. As such, we have the most confidence in this part of the NO_x inventory. The text was altered to clarify as follows:

> The emissions included data from point sources equipped with continuous emissions monitoring systems (CEMs) that measure SO_2 and NO_x emission rates and other parameters daily, mobile emissions processed by the Mobile6 model, and meteorologically adjusted biogenic emissions from Biogenic Emission Inventory System (BEIS) 3.13 all specific for the year 2004.

P6481, L4: Prior to Hudman et al. 2007, Cooper et al. 2006 published a report of the extensive NO_x 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.

The reference to Cooper et al., (2006) was added to this section of the manuscript.

P6481-6484: A spatially uniform increase in NO_2 columns was used to account for missing lightning NO_x 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 NO_x source)? Even if such an analysis is beyond the scope of the paper, please comment on how a spatially inhomogeneous upper tropospheric NO_x source might be included in your analysis.

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In this manuscript, we tried to focus more on the methodology of the inverse, however, we have done some of work the reviewer suggests. Unfortunately, simply adding NO_x emissions aloft (from any source) is not sufficient to increase NO_2 column concentrations. Currently, CMAQ, and other models, tend to terminate NO_2 in the upper layers too quickly. Our experiments with adding lightning NO_x production based on flash measurements confirmed this occurrence and we are currently concentrating on improving the chemistry aloft to resolve this issue. Certainly, spatially resolved sources would improve this type of 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.

Macon was used as an example, but the Mississippi region also demonstrates the influences of other uncertainties, aside from emissions, on the results of the inverse. The text was modified to reflect this suggestion as follows:

> In the "Macon" and "Mississippi" source regions, adjustments to the inventory are outside the specified uncertainty of the emissions inventory (factor of two).

*P6470, L8-10: suggest rewrite as follows: *as constrained by observations of NO_2 column densities derived from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) satellite instrument.**

Done.

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

This sentence was removed completely to address another reviewer comment.

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

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Done.

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

Changed as suggested.

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.

Yes, this factor was chosen arbitrarily to be some small fraction of the diagonal terms. The following text was added to this section of the manuscript to explain:

> The off-diagonal elements of the covariance of error matrix are difficult to estimated, and in this application were set to be a fraction (10%) of the average of the corresponding diagonals (Eq. 5).

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

Done.

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

Done.

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

Done.

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

Done.

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

Done.

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

Done.

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.

The figure was modified to include source region names.

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