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Atmospheric Chemistry and Physics Discussions

ACPD

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Interactive Comment

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 #2

Received and published: 8 May 2008

This is an interesting paper that addresses the relevant issue of how to accurately infer NOx emissions from satellite NO2 measurements. The authors discuss their inverse modelling system, demonstrate that it works by doing an end-to-end test using pseudo-observations, and finally apply the method to infer NOx emissions from SCIAMACHY NO2 retrievals for the southern United States during the summer of 2004. They make a convincing case for the need to include realistic lightning NOx in their chemistry-transport model in order to arrive at reliable surface NOx emission estimates. This is an important conclusion, and to my knowledge the first time that the systematic error

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Interactive Discussion

Discussion Paper

resulting from a poor simulation of background NO2 is quantified based on a set of state-of-science NO2 observations in the upper troposphere.

On the other hand, the paper reads somewhat like a collection of missed opportunities. The paper could be strengthened if the authors make matters more practical and quantitative. Below are some examples, and I suggest the authors take these into account in their paper:

* The difficulty in data assimilation is often in attributing realistic errors to the model and the observations. The authors discuss the effects of a range of combined measurement and a priori emission errors (Fig. 5) for the pseudodata case, but avoid stating what exact numbers they used in their case study and why. I think these numbers and some justification thereof should be given.

* The pseudodata analysis suggests that border regions and boundary conditions have minimal influences on the inversely modelled NOx emissions within the region of interest. This is an important result, and I'm wondering why the authors do not include it in the abstract. Furthermore the lack of discussion of this finding is puzzling. The result seems to be specific for summertime southeastern US (short chemical lifetime, stagnant weather), and it needs to be discussed in that context.

* The authors do not discuss the impact of the small number of SCIAMACHY observations (3-10 over the whole period) on their results whereas they could easily have done so. For instance, the pseudodata analysis was done for 1 August 2004 with base-case model simulations as pseudo observations. Such a test is useful, but not at all representative for the inverse method with real SCIAMACHY data. A test with pseudo observations sampled as SCIAMACHY observations would specify to what extent the observations contribute to finding the solutions in your real data. Similarly, the diagonal elements are set to 0.5 1015 molec. cm-2 (P6478, L26-27) to test the pseudodata analysis, and I'm wondering why the impact of more realistic observational errors has not been tested here.

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* Why have the authors chosen such large (200x300 km2) source regions? This is hardly taking advantage of the high resolution SCIAMACHY data.

Then I have concerns about a number of methodological issues: The authors assume that atmospheric NO2 concentrations at 10:00 reflect the NOx emitted in the previous 16 hours only (P6478, L4-7). This suggests that NOx emitted in the afternoon rush-hour does not contribute in any way to NO2 observed the following morning at 10:00 am. But some afternoon rush-hour NO2 will live through the night and may contribute to NO2 observed the next day at 10:00 am, especially downwind of strong sources. I understand that summertime NO2 has a short (daytime) chemical lifetime of 2-4 hours, but at night the chemical lifetime is longer. I think the authors should justify the implicit assumption that 10:00 am NO2 concentrations are unrelated to afternoon emissions. Related to this issue, I think it is important to include some discussion on the timing of the NOx emissions in CMAQ.

Furthermore I suggest the authors discuss the representativity of the surface NO2 measurements in more detail. How representative were the SEARCH sites for the average concentrations simulated for the 36 x 36 km2 CMAQ grid cells? Why were the surface measurements averaged over the daytime concentrations rather than sampled at 16:00 UTC as was done in the CMAQ-SCIA analysis?

In the Abstract, reasons for the underestimation of NO2 columns are mentioned. The authors convincingly point out that the lightning NO2 is likely too low in the CMAQ model, but also mention "a short modelled lifetime of NOx aloft" as a likely reason but without substantiating this in the paper.

Minor comments P6471, Ll28: please spell Müller not Muller.

P6472, L1: please spell Quélo, not Quelo. Although Quélo's is an interesting paper, it doesn't use SCIAMACHY or any other space-based observations as the sentence now suggests. Please update.

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P6472, L1: it would be appropriate to also cite the work by Blond et al. (JGR, 2007), Y. Wang et al. (GRL, 2007) and Konovalov et al. (ACPD, 2008) here. These papers use high-resolution CTM simulations, SCIAMACHY/OMI and surface NO2 to better understand air pollution on the regional/urban scale.

P6472, L6: please provide a citation to DDM-3D here.

P6475, L10: I don't get this. At eq. (2), it is stated that N includes not only observation errors but also model uncertainties. This sentence suggests we're only dealing with observation errors here. Please clarify.

P6475, Eq. (6): I think the right-hand side of the expression should be squared since it is a (co)variance matrix. Furthermore, there is now inconsistency with Eqs. (4), (5) and P6478, L27.

P6475: I feel the paper would be strengthened if the authors give a range of numbers for UE,m and Uobs, and some justification for these estimates.

P6477, L14-16 ("The inverse was ... data as Xmod"): I don't understand what this sentence should tell us. Could you please clarify?

P6478, L20: I think the reference should be to Eq. (6) here, not (5).

P6479, L14: I think the authors also want to refer to Eq. (6) here.

P6479, L16: there is no Eq. 5a to refer to.

P6479, L24-25: I think it should be stressed here that Fig. 5 relates to the Atlanta case only.

P6481, L5-11: this raises the question if and how lightning NOx is simulated in CMAQ. Please describe the (lack of) of LNOx simulation. P6481, L13: Konovalov et al. have not found a systematic bias between satellite observation their model simulation over Europe. They just report that NOx emissions from lightning are not included in CHIMERE, and state that this likely leads to an underestimation of the NO2 column of

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at most 0.08 1015 (not 0.8 1015 as the authors suggest here). Their deficiency is thus more than an order of magnitude smaller than the 1.07 1015 reported here. Please clarify.

P6482, L15-16: I suggest the authors provide some more information on the surface NO2 measurement technique, the Hansen et al.-paper was not readily available to this reviewer.

I recommend this paper for publication in ACP if the authors revise their manuscript according to the suggestions and criticisms given above.

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

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