**Atmospheric
Chemistry
and Physics
Discussions**

Interactive
Comment

Interactive comment on "Global NO_x emission estimates derived from an assimilation of OMI tropospheric NO₂ columns" *by* K. Miyazaki et al.

Anonymous Referee #2

Received and published: 9 February 2012

The authors have used a local ensemble transformed Kalman filter (LETKF) for inverse modeling of NOx emissions using NO2 observations from OMI. As the authors noted, this is the first use of the LETKF for inverse modeling of NOx emissions. It represents a significant step forward from previous inverse modeling approaches used for quantifying NOx emissions, such as the mass balance approach. The LETKF offers an effective means for characterizing model errors in the inversion, which is critical for obtained reliably source estimates. Since NOx plays a key role in the oxidative chemistry in the troposphere, changes in NOx influences the abundance of other tracers such as ozone, and the LETKF provides a useful way of capturing the correlation between species in the inversion. Because it represents an innovative approach for inverse

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

modeling of NOx emissions, I believe that the manuscript is suitable for ACP. However, I do have some concerns, described below, which I believe must be addressed before the manuscript will be suitable for publication.

Major Comments

1) The limited validation conducted with in situ aircraft data does suggest that the inversion is improving the modeled NOx distribution. However, I think that in the absence of more extensive validation with in situ data, it would be extremely important for the authors to put their estimated NOx emissions in context with what is in the literature. For example, Hudman et al. (JGR, 2007) suggested that power plant and industrial emissions of NOx in North America decreased by 50% between 1999 and 2004. How consistent are the OMI derived NOx estimates with those from other studies, such as the Hudman et al. analysis?

2) The vertical profile comparison (Figure 11) shows that the model is significantly underestimating free tropospheric ozone. My guess is that this is linked to an underestimate of lightning NOx emissions (LNOx), which could bias the surface source estimates of NOx. In particular, I wonder to what extent could this account for the inferred increase in NOx emissions in the eastern USA in the inversion? Indeed, when the authors reduced the LNOx source in the model, the a posteriori NOx emissions were much larger. I believe that it would be helpful for the authors to compare their regional LNOx estimates with those from other studies such as Hudman et al. (JGR, 2007), Sauvage et al. (ACP, 2007), and Jourdain et al. (ACP, 2010).

3) The authors invested significant effort in evaluating the performance of the forward model and the LETFK assimilation system, which I appreciate, since this is the first application of the LETKF for inverse modeling of NOx emissions. However, there are a few key parameters in the assimilation system for which the values were specified in an ad hoc manner without explanation. For example, on lines 16-18 on page 31537, the authors state that the analysis errors were inflated to 30% of the initial standard

deviation, based on sensitivity tests. What tests? What metric was used in these test to determine that 30% was the best value? Similarly, on line 1, page 31540, there is no explanation as to why a 15% error correlation was used in generating the super-obs. It is clear that there was quite a bit of tuning of the assimilation system and it would be helpful if the authors gave the reader a better sense as to why the different parameter values were selected for the standard inversion case.

Minor Comments

1) Line 10, page 31525: Please change "formations of HNO3" to "formation of HNO3"

2) Lines 2 – 5, page 31526: I don't understand what the authors are saying here. How is the total column more closely related to the area average emissions than surface data? With the column data you have the contribution from the stratosphere and upper tropospheric sources, such as lightning.

3) Line 28, page 31526: Please add "us" after "allow"

4) Line 17, page 31528: The averaging kernel is not important in the retrieval. In fact, it is not used in the retrieval. It is a by-product of the retrieval that enables one to assess the sensitivity of the retrieval.

5) Line 11, page 31537: Please change "out setting" to "our setting".

6) Line 27, page 21537: Why did the authors add noise with a magnitude of 4% of the initial spread? This 4% seems ad hoc. Can they justify this value?

7) Lines 1, page 31540: On what analysis is the 15% correlation based?

8) Equation (11), page 31540: I don't quite understand the role of alpha here. What does alpha = 0 imply physically?

9) Figure 2: Are the panels with the difference maps showing the differences between the data and the model with the averaging kernels? It is not clear from the caption. Using the untransformed model in the difference maps would be less useful.

ACPD

11, C15204–C15207, 2012

Interactive Comment

10) Line 4, page 31544: Europe looks good, contrary to what is stated in the text.

11) Figure 4: What are the error bars for the satellite data? Since this is an average over a large region, what is the variability around the mean value?

12) Lines 21-25, page 31544: The improvement with the diurnal variability scheme is not obvious to me.

13) Lines 10-14, page 31546: Is the better performance with the super-obs just due to the fact that you are averaging a large number of retrievals in each gridbox, and as a result the retrieval noise is significantly reduced? Since the precision on an individual retrieval is not very high, one could image that the noise from each retrieval could cause the inversion to produce noisy increments.

14) Lines 1-6, page 315550: I am surprised that the inversion has sufficient sensitivity to optimize the ship emissions. Over the oceans the NO2 column is low, and the stratospheric contribution to the column will be higher than over a continental source region. It would be helpful to see the results of an OSSE showing that the inversion is indeed sensitive to these emissions, given the specified measurement and model errors.

15) Figure 12: It is not clear what the different lines represent in the scatter plots in panel (b) and (c). Is each line a linear fit to the data points of the same color as the line? Given the significant scatter in each panel, I wonder how meaningful is the linear fit?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 31523, 2011.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

C15207