

***Interactive comment on “Global NO<sub>x</sub> emission estimates derived from an assimilation of OMI tropospheric NO<sub>2</sub> columns” by K. Miyazaki et al.***

**Anonymous Referee #1**

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This paper reports estimates of global NO<sub>x</sub> emissions based on data assimilation of the DOMINOv2 retrievals of tropospheric NO<sub>2</sub> columns from the OMI instrument in the CHASER global model. To this purpose, a data assimilation system based on the local ensemble transform Kalman filter (LETKF) is developed and implemented (Sect. 2.3) together with a super-observation approach (Sect. 2.3.3). Comparisons with an older version of OMI (v1) NO<sub>2</sub> dataset and the SCIAMACHY NO<sub>2</sub> data are carried out, and the assimilation output is validated against vertical profile measurements from the INTEX-B (US-Mexico, March 2006) and DANDELIONS (Cabauw, September 2006) campaigns. The scope and relevance of the article is of interest to merit publication in *Atmospheric Chemistry and Physics*. The study is thorough. Different ways to validate

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the results and to perform sensitivity assimilations are considered. The results look correct and the conclusions drawn are generally robust.

However, the readability of the article is seriously hampered by its excessive length. Discussion could be more synthetic and insightful. At many instances, it is either limited to a simple reporting of the content of the figures with too much detail (e.g. Sect. 3.2, 3.3, 5, 6) or it consists of commonplace statements which do not bring essential information. I strongly recommend that authors make an effort to increase the conciseness of their text and remove speculative/unproved arguments.

The manuscript can be accepted for publication only after the following points are adequately addressed and elucidated.

Comments:

1. Could the emission increases over several regions (e.g. China, Eastern US, S. Africa) inferred by the assimilation (cf. Table 3) be driven by the general underestimation of tropospheric NO<sub>2</sub> columns in the CHASER model as reported in the intercomparison paper of van Noije et al. 2006?
2. To assess the validity of the assimilation results the a posteriori regional/global emissions should be compared with previously reported top-down estimates.
3. Besides introducing the basic LETKF formulation, the article does not go deeply enough in the analysis of the individual steps needed to obtain the emission updates. Furthermore, more details are needed on the advantages of the LETKF method over EnKF, on the localization strategy and the covariance inflation. In addition, please explain also how the perturbations  $X^b$  (p. 31534, l. 14) are defined. In Equation 4, the covariance inflation parameter  $\Delta$  as well as the particular type of inflation assumed here should be defined. In the same formula,  $I$  is the identity matrix, so it should be in bold roman style.

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4. Section 3.2 on the seasonal variation of the NO<sub>2</sub> columns is too long and can be easily shortened. Some of the sentences (e.g. p. 31543, l. 22-24, l. 27-28) are commonplace and should be omitted. Further, it is not true that there is an obvious underestimation in winter in Europe (as stated in page 31544, l. 3), at least when we compare with DOMINOV2 data. Please correct the text (p. 31544, l. 4).
5. The first paragraph of Section 3.3 could be shortened or even omitted. In p. 31544, second paragraph, it is stated that the use of diurnal profiles in CHASER improves the model-data agreement globally. However, the agreement worsens over Central Africa (Fig. 4b) after application of the assumed biomass burning profile. The authors should first give more details about this profile and make a point on this in the main text to explain the model behaviour in Fig. 4b.
6. Section 5.1 is too long and contains much self-explained information. The authors do not make any point on the difference between the seasonality of emissions over Europe and the Eastern US (Fig 7, top panels) and on the much higher NO<sub>x</sub> emissions derived by the assimilation over the Eastern US. This latter result seems to be at odds with what we know from EPA trends of NO<sub>x</sub> emissions over the US. Some discussion is needed.
7. In Section 5.1 (p. 31547, l. 20) the authors state that the REAS inventory largely underestimates the NO<sub>x</sub> emissions over Eastern China in 2005, as commonly revealed by van Noije et al. (2006). But van Noije et al. (2006) do not make an evaluation of the REAS inventory. This should be corrected. On the other hand, it should be specified how the extrapolation of emissions is performed. In the case of the REAS inventory, an extrapolation is not necessary since 2005 and 2006 REAS emissions are already available online. In addition, the GEIA emission inventory for soils cannot be considered as a "latest" inventory, as mentioned in p. 31547, l.26. Also in p. 31548, l. 11, "the a priori and the latest ones" should

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- be changed.
8. In Section 5.1 (p. 31550, first paragraph) the statement about oceanic NO<sub>x</sub> data is misleading since the Boersma et al. (2008) OMI data are not the same as the DOMINOv2 product used in the assimilation. To make a meaningful statement, you must compare DOMINOv1 and DOMINOv2 over the oceans.
  9. In Section 5.3, p. 31553, l. 9-10, it is stated that “above the PBL, the NO<sub>2</sub> concentrations decrease with height, primarily because of the NO<sub>2</sub>/NO ratio, which decreases with temperature”. The main reason for the decrease is the relatively short lifetime of the NO<sub>x</sub> family, due to the NO<sub>2</sub>+OH reaction. Change in the NO<sub>2</sub>/NO ratio play only a minor role.
  10. p. 31531, l. 32 : correct “metrological” in this sentence
  11. p. 31533, l. 10 : correct “maxima emissions”
  12. p. 31534, l.1 : missing reference to Whitaker and Hamill
  13. p. 31537, l. 11 : correct “in out setting”
  14. p. 31541, l. 21 : “occurs in relatively fewer observations”, do you mean “leads to relatively fewer observations”?
  15. read names “van Noije” and “van der A” throughout
  16. p. 31542, l. 11 : “the CHASER” remove “the”
  17. read “molec./cm<sup>2</sup>” instead of “mol/cm<sup>2</sup>” throughout
  18. p. 31549, l. 18 : “of greater” remove “of”
  19. p. 31552, l. 27 : “below 950 hPa”, read “at pressures higher than 950 hPa”, similar correction in l. 28.

20. p. 31553, l. 6 and 8 : read "increase by a factor"

21. p. 31553, l. 17 : "Cabauw...100 km", poor phrasing

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

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