

***Interactive comment on “Assimilation of OMI NO<sub>2</sub> retrievals into the limited-area chemical transport model DEHM (V2007.0) with a 2-D OI algorithm” by J. D. Silver et al.***

**Anonymous Referee #2**

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In their paper the authors discuss an assimilation of satellite observations of NO<sub>2</sub> columns with an OI scheme and a state of the art chemistry transport model. Extracting information on air pollution levels from satellite observations is a very relevant topic. The paper is generally well written. However, I have a list of seven major points of concern, detailed below. Because of this I judge that a major revision is needed, which should address all these points before the paper can be accepted for publication.

General remarks:

The authors did not make full use of the OMI data product. Averaging kernels provided in this product have not been used. Also the error estimates available in the product

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have been used only in experiment 4 (5), which apparently is not even the optimal choice (which is exp 3 with constant errors). The vertical sensitivity (or averaging kernel) of the measurement technique is strongly height dependent, and residual clouds have a strong impact on what can be observed from space. In particular OMI is extra sensitive to NO<sub>2</sub> above the PBL, and the amount of free troposphere NO<sub>2</sub> modelled by DEHM should be investigated, reported and compared to other studies (see references). Why have the kernels not been used? The implementation of the observation covariance R is not clearly explained, see Table 1. What does  $R_{ij} = 1$  mean? How are exp 4 and 5 implemented?

The authors mention that negative estimates of the tropospheric column are excluded. However, such negative values in the OMI product seem to represent the uncertainty in the retrieval in cases where the amount of NO<sub>2</sub> is less than the error bar. Excluding those negative numbers will generally result in positive biases in clean areas. Does this explain part of the positive adjustments made in the assimilation process?

The lifetime is an important issue. If (in Summer) the NO<sub>x</sub> lifetime is only a few hours, the impact of data assimilation will be lost within the same few hours. Since OMI is only available once per day, how can the authors claim that OMI can be used for reanalyses using the OI technique? Fig. 8 seems to suggest longer lifetimes than suggested by the introduction (between 3 and 13 hours). Please explain this difference. Also, the difference between the reference run and exp 3 seems to be tiny (the NO<sub>2</sub> curves are basically on top of each other). Is this correct? It seems incompatible with the considerable adjustments seen in figures 4-6. What is the mechanism to keep the NO<sub>x</sub> adjustment information in the model for 24-48 hours.

The Hollingsworth and Lonnberg approach to estimate model and observation contributions to the error covariance is not simply applicable to satellite observations. The main assumption in this approach is that errors in the measurements are uncorrelated in space, which is often a reasonable assumption for well separated surface stations, but generally does not apply to satellite data products, given their sensitivity to e.g.

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cloud fields and surface scattering properties. When errors are correlated the estimate of the background error weight  $\theta_b$  (eq.2) is no longer meaningful. The error correlation above the intercept could still be due to the model, e.g. wrong local (within one grid cell) emissions. On the other hand, part of the spatial correlations could be due to the satellite observation errors.

The authors do not present a-posteriori validation for their OI approach. Are the observed differences (observation minus forecast) compatible with the covariance matrices used ( $\chi^2$  test)?

It seems that the main impact of the assimilation is a correction of an overall negative bias in the model. Would it be better to replace the OI scheme by a bias-correction scheme (described in papers by e.g. Dick Dee)?

The authors compare with profile information based on ozone sondes. I fail to see why this is very relevant for this paper. Instead it would be good if the authors can do some "confidence building" to show that the NOx/NO2 profile in the model is reasonable up to the tropopause, by comparing for instance with other models and/or with available observations. Just showing the model profile would already be of use.

More detailed remarks:

p312-313, intro: The part on NOx chemistry and impact on health is basic background knowledge and can be summarised in a few lines with references. I would have expected this part at the beginning of the introduction. The lifetime seasonality remark is relevant and should be kept.

p314, l26: Negative estimates of the tropospheric column are not an artifact of the retrieval, but represent the uncertainty in the retrievals.

p315, l12: "if any"

p315, l14: "unreliable" what is the criterium?

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p316: Have the DEHM free troposphere (NO<sub>x</sub>) concentrations been validated? Are relevant processes for the free troposphere (lightning, convection) included in the model?

p317: Eq 2: B has the dimension of x-squared, so the column concentration squared. Is this dimension included in  $\theta_b$  ?

p318, eq 3: ? Please provide additional motivation for this form. Why is model-forecast error correlation length depending on the number of observations? From a theoretical point of view it should be completely independent of the density of the newly added observations, but there can be a (often weak) dependence on the observations of the previous day. Given the short residence time of NO<sub>x</sub> I would expect this dependence to be very weak.

p318, l10: Why would the length scale for ozone be relevant for NO<sub>x</sub> ?

Sec 2: Is the model bias-free compared to OMI: long-range correlations will mess up the length scale parameter estimates. Has the estimate of the innovation correlation been debiased?

p318. l10: See my general remark on the Hollingsworth-Lonnberg approach.

p318, l25: Why is a binning to grid points needed? The procedure to compute correlations is described very extensively (six steps). I would suggest to summarise this in 1-2 lines.

p319, eq 4: Please explain the  $r^2$  normalisation in this formula. Does this account for the increasing number of pairs at larger distances?

p320, l22:  $R_{ii}$  should scale like the observation error squared. Has this been done, or is  $R_{ii} = \sigma$ , which would not be consistent with the Kalman filter equations. Please be more explicit on the exact form of  $R_{ii}$  in exs 3, 4 and 5. What does  $R_{ii}=1$  mean?

p320, l27: Again, the form  $y/\sigma$  has no dimension, while  $R_{ii}$  should scale like the

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observation error squared. How is this implemented?

Sec 3.2: Are the EMEP observations representative for the grid cell of DEHM? Please discuss this issue. Or would one expect offsets? NO<sub>2</sub> surface measurements often contain other nitrates like HNO<sub>3</sub>, PAN. Has this been corrected for?

p321,322: It would be useful to provide a short explanation how R2, NMSE and FB are defined.

Sec 3.4: replace "ozonosondes" by ozone sondes or ozonesondes (several times in text)

p322: Why are De Bilt and Legionowo chosen?

Section 4. Sections 3 and 4 should be merged into one section, e.g. combine 3.2 with 4.1 etc. . . The split is artificial and is not helping the reader.

p323, 115: It is a bit disappointing that constant errors seem to perform better than using the error bars in the OMI product. Adding more information should help.

325, sec 4.4: The relaxation time of 2-3 days is long compared to other lifetime estimates. See above.

325, bottom: It is not clear to me why the lower correlations are resulting from a spread of information to cloudy parts. Please explain more clearly. There can be multiple other reasons.

326: The NO<sub>2</sub> profile shape is kept constant. I do not see how this can be validated with ozone. Please provide a discussion on the quality of the NO<sub>2</sub> model profiles (see above).

326, 121: The lower background weight: See remark on the Hollingsworth and Lonnberg approach above.

327, 120: It would have been good to discuss the forecast impact in more detail, as was

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done in Wang.

Table 2 and 3: The bias of the model compared to OMI and surface observations is quite large (38%). It seems that a bias correction scheme could be more appropriate, see general notes above.

Fig 2: Density plot: it would be better to use colors and include a log-scale legend to provide actual counts. In this way one can judge the spread of values in a better way.

Fig 3: There is apparently very little seasonal variability in model? Why? As mentioned in the intro the lifetime strongly depends on the season. What does "calculated" refer to, i.e. is this the reference run or one of the assimilation runs ?

Fig 4: How can there be such large increases at spots where no OMI data is available (grey)?

Fig 6: Is this consistent with fig 3? There seem to be large increases also in spring-summer in the data assimilation. The simulation in Fig 3 seems to suggest that the bias in spring-summer is small.

Fig 7: Could be more clear if the horizontal scale is blown up (lower stratosphere is not so interesting for this paper) so that lines in the lower troposphere are more clearly separated.

Fig 8: Is the relative difference consistent with the lines? The two model simulations seem to overlay almost perfectly, while I would have expected a significant difference?

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