Reply to Reviewer #2

Review of "New concepts for the comparison of tropospheric NO2 column densities .."
by Shaiganfar et al.

In this manuscript, the authors report on a systematic comparison of a large number of car MAX-DOAS measurements of tropospheric NO2 columns in Paris to CHIMERE model results and OMI satellite observations. They successively apply a number of modifications to the data (rotation of model data to account for errors in the met-fields, use of high resolution model a priori profiles in the OMI observations, and application of spatial smoothing to the car MAX-DOAS measurements) and discuss the impact of these modifications. They present a detailed comparison of the different data versions between each other and finally use the model data as transfer to correct for the difference in spatial resolution between satellite and car MAX-DOAS data.

The paper is well written, clearly structured and reports on a nice and very thorough comparison. The methods presented are relevant for other similar studies and validation of model and satellite data with local data in general and fits well into the scope of AMT. I therefore recommend publication after minor revisions as suggested below.

Author reply:
We thank the reviewer for the positive assessment and the useful suggestions. We addressed almost all of ths suggestions as described in detail below.

General Comments

• Improved correlation is used in several places as indicator for better orientation of model data, better representation of spatial scales, and better resolution of NO2 gradients when using high resolution a priori profiles. While I think that this is a valid approach, the authors need to acknowledge that a) an improvement in correlation is to be expected if a parameter is varied until the correlation is improved (rotation, smoothing) even in a random data field and

Author reply:
We agree and changed in section 4.1 the original text:
'The applied rotations cause a substantial improvement of the correlation coefficients. Also the slopes of the regression lines increase while the y-axis intercepts decrease.'

Into:

'The applied rotations cause a substantial improvement of the correlation coefficients. While such an improvement has to be expected (and was the criterium to determine the rotation angles), it is interesting to note that also the slopes of the regression lines increase while the y-axis intercepts decrease.'

b) that the use of CHIMERE NO2 profiles would improve the correlation with CHIMERE columns even in the hypothetical case of a homogeneous OMI NO2 field (I would expect the correlation to increase from close to 0 to close to 1 in such a case).

Author reply:
We agree and changed in section 5.1 the original text:

'Here it is important to note that, although the modified OMI data are partly dependent on the CHIMERE profiles, it still makes sense to compare the modified OMI data to the tropospheric NO_2 VCDs extracted from the CHIMERE data, because the modified OMI data only depend on the relative shape of the simulated NO_2 profile, but not on the absolute value of the tropospheric NO_2 VCD.'

into:

'Here it is important to note that, the modified OMI data are partly dependent on the CHIMERE profiles. In an extreme case, for example, the application of CHIMERE profiles to a hypothetic homogenous (non-zero) OMI NO_2 field would cause a deceptive high correlation between OMI and CHIMERE data. Nevertheless, it still makes sense to compare the modified OMI data to the tropospheric NO_2 VCDs extracted from the CHIMERE data, because the modified OMI data only depend on the relative shape of the simulated NO_2 profile, but not on the absolute value of the tropospheric NO_2 VCD.'

• The rotation of the CHIMERE field is a nice approach and the improvement quite impressive for the car MAX-DOAS but the divergence between the OMI and car MAX-DOAS angles is a bit sobering and might be indicative of artificial improvement of the agreement through this "free parameter". In this context one could also speculate that the intrinsic rotation of the OMI pixels relative to the regular CHIMERE grid might introduce ambiguities.

Author reply:
In section 4.1. we already discussed the probable reasons for the disagreement between rotations determined with either car-MAX-DOAS and OMI:

'The rather low correlation is probably caused by the fact that the comparisons of the model data with both observational data sets are made for different times and locations. In particular the comparisons versus OMI observations are performed for a much larger area (see Figs. 3 and 4).'

Author reply:
In the revised version we changed the text (in section 5.2):

'Like for the comparison with the car-MAX-DOAS measurements, for most days the rotation of the CHIMERE data leads to an improvement of the correlation coefficients. However, for the slopes and y-axis intercepts only small changes are found.'

into:

'Like for the comparison with the car-MAX-DOAS measurements, for most days the rotation of the CHIMERE data leads to an improvement of the correlation coefficients (as has to be expected). However, the improvement of the correlation coefficient is smaller than for the comparison with the car-MAX-DOAS measurements. Also, for the slopes and y-axis intercepts only small changes are found. Both findings indicate that the determination of the rotation is less well constrained by the OMI observations compared to the car-MAX-DOAS measurements. This can be explained both by the much coarser resolution of the OMI data and the frequent gaps due to clouds. Since the spatial resolution of the CHIMERE data is much finer than the OMI resolution, the shape of the OMI ground pixels has no significant effect on the determination of the rotation angles, as CHIMERE data is re-sampled to the OMI pixel extent.'

• The smoothing approach to account for the effective spatial resolution of CHIMERE is a good idea but again I see the risk of compensation of other problems in the model (emissions, lifetime) by the free parameter.

Author reply:
This possible reason was already discusseed in the original version of the manuscript (in section 4.2):
'Interestingly, the optimum horizontal smoothing kernels are significantly larger than the spatial resolution of the CHIMERE data (3 km). This result was unexpected, and the potential reasons for the need of an additional smoothing are not completely clear. Probably some atmospheric process(es) relevant for the dispersion of the NO_2 plume are not sufficiently well represented in the model. Such processes might include atmospheric mixing but also the characteristic times of chemical reactions. Alternatively, also the spatial distribution of the emission sources used in the model simulations might be too coarse, or their spatial emission distribution might be imperfect at small scales of some kilometers. In all these cases, smoothing of the spatial scales reduces model errors and improves comparison with observations.'

We feel that this point of the reviewer was already well covered by that discussion. However, according to the next reviewer comment, we added the following sentence:

'In addition sampling effects and numerical diffusion might also contribute.'

Also, the correlation maximum as shown in Fig. 11 is quite shallow, making the result less robust than what one would hope for. Considering the native model resolution of 3km, sampling issues and numerical diffusion in the model might also be relevant for a result of 5 km optimum smoothing.

Author reply:
See reply to last reviewer comment (above)

Detailed Comments

P2441, l22: two => to
Author reply:
Corrected (also 'verticaol' => 'vertical' in the same line')

P2441, l22: Sentence with Deguillaume et al. reference not really needed as VOC sensitivity is not taken up again in the paper

Author reply:
This sentence and reference had been added to the revised ACPD paper following the request of a referee to give information of former modelling studies dealing with NOx in the Paris region. We can indeed give a more specific reference and changed the text (and reference) accordingly :
"Deguillaume et al. (2007) used urban NO and O_3 concentrations from the AirParif network to constrain urban and plume ozone concentrations in a Bayesian Monte Carlo framework. For summers 1998 and 1999, they found good agreement between urban background NO concentrations (12.8 ppb, average over 5 sites) and simulations with the CHIMERE model (12.6 ppb)".

P2441, l27: Comparison is not to top-down inventories but rather to measurements meant to be used for such inventories

Author reply:
We replaced 'top-down inventories' by 'car-MAX-DOAS measurements around emission sources'

P2442, l9: synergistic use of what?

Author reply:
The sentence is reformulated to: 'We make synergistic use of the different data sets by combination their specific advantages.'

P2443, please add some details on NO2 fit such as fitting window, background spectrum used and approach to measurements blocked by buildings etc.

Author reply:
We added the following information to section 2.1:
'The spectral range from 420 nm to 460 nm was used, and in addition to the NO_2 cross section (294 K, Vandaele et al., 1998) also those of H_2O (290 K, Rothman et al. 2005), CHOCHO (Volkamer et al., 2005), O_3 (343 K, Bogumil et al., 2003), and O_4 (296 K, Hermanns et al., 1999) and a synthetic Ring spectrum (Wagner et al., 2009) were included.. A daily measurement in zenith direction is used as so called Fraunhofer reference spectrum. Measurements for which the light path was blocked by trees and buildings were sorted out by appling a threshold to the magnitude of the residual of the spectral fit.'

P2444, l13, not sure if information on GPS systems used is relevant

Author reply:
we removed the detailed information about the GPS system

P2446, l25, sentence appears twice
Author reply:
The (second) sentence was removed.

P2450, points (d) and (e) do not fit into the list of "corrections to the original data"
Author reply:
points (d) and (e) have been incorporated in the main text.

P2451, l5, These discrepancies => This discrepancy
Author reply:
corrected

P2454, section title, Influence of => influence of applying
Author reply:
corrected

P2459, l25, measurements => data
Author reply:
corrected

P2460, l13, I find this assignment of uncertainties to CHIMERE very arbitrary!

Author reply:
We added the following explanation to the text:
'Of course, the choice is arbitrary, but it is only technical in order to perform the linear regression. The exact value of the assigned uncertainties only little affects the results of the fit. Unambiguously assigning uncertainties to modelling results is difficult, because for instance differences to observations are always affected by representativness and measurement errors.'

P2464, l24, Another possible explanation for the observations would be a misplacement of emissions around Paris – to my knowledge, there was a problem in some emission inventories that put all emissions to the city and nothing to the suburbs (sorry, I have no reference for that: : :)

Author reply:
This possibility could also be envisioned, and a corresponding sentence is added in 'Summary and Outlook':

"A spatial misplacement of emissions in the Paris region, in particular as a function of the distance from Paris center, is also a possible error source, as has been shown by comparing NOx emission fields in and around the Paris agglomeration from different emission inventories (Petetin et al., 2014).

Figure 18: Something seems wrong with that figure – why is the maximum of the OMI data smaller in the modified data set?

Author reply:
We corrected Fig. 18.