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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Comparison of OMI NO_2 tropospheric columns with an ensemble of global and European regional air quality models" *by* V. Huijnen et al.

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General comments:

We thank reviewer#2 for his valuable comments to our manuscript, which helped to formulate our objectives, analyzes and conclusions more accurate.

RC1-G: Despite the similar setup the models often show large differences. A clear weakness of the study is that the reasons for these discrepancies remain mostly unclear and are not further investigated. I acknowledge though that the specific setup of the models in an operational environment did not allow for an in-depth analysis of such differences which would require further sensitivity studies. Nevertheless, this is a

significant drawback that needs to be considered in future model evaluation studies.

AC1-G: The focus of our manuscript is an evaluation of the GEMS-RAQ system as a whole, rather than the individual model performance. This is addressed more clear in the revised manuscript. As suggested by rev#1 we have created an ensemble median. Additionally we also analyze the deviation of individual models compared to this reference, but we agree that fully satisfying explanations for differences in model behavior cannot be achieved in this context, where the contributing models vary in many respects.

RC2-G: The bulk of the regional models actually performs quite similar (see e.g. Figure 7 and 9) with MATCH and SILAM often showing up as negative and positive outliers, in particular during summer. The CHIMERE model simulates very low concentrations over Eastern Europe which suggests a problem in the implementation of the TNO emissions inventory, but I don't think this is mentioned anywhere in the paper. I think these facts should be stated more explicitly, rather than trying to defend individual model problems.

AC2-G: In the revised manuscript we make statements on individual model shortcomings, like the problems with the emission inventory in MATCH and MOZART-IFS more explicit. They are now mentioned in the model-description, see also rev#1, AC2-G. The low concentrations in CHIMERE over eastern Europe indeed suggests a problem with the implementation of the emissions, similar to what was found in the MATCH model. However, this has been double-checked, and so far no implementation error was discovered. Therefore we did not include such a statement in the manuscript. However, a recent version of the CHIMERE model where the TNO inventory is replaced by the EMEP inventory does not show similarly low concentrations for winter 2010 as for winter 2009 where the TNO inventory was used. On the other hand, the EMEP inventory is not significantly different for eastern Europe compared to TNO, and the difference cannot explain the differences in modeled NO2. This again suggests an implementation error with the TNO emissions inventory in CHIMERE.

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As also noted by the reviewer, from several analyses in the original manuscript it can be observed that the SILAM model showed relatively large background concentrations compared to the other models. A clear explanation was missing so far. Because of its exceptional behavior it was decided to investigate this in more detail, by performing three sensitivity studies: (1) replacement of the TNO inventory with the EMEP inventory (2) an alternative vertical diffusion parameterization, which resulted in substantially lower vertical mixing rates, combined with an increased dry deposition velocity. (3) The replacement of the gas-phase chemical mechanism with the basic version of CBM4, Gery et al. (1989).

The simulations with different emission inventory with coarse spatial resolution (50km) but the same country totals showed minor differences in terms of the background column loads but sharply reduced the peaks over the major cities.

The change in vertical diffusion resulted in almost proportional decrease of the column values everywhere due to increase of sinks – both via chemical reactions and via dry deposition from the lowest model layer where the bulk of the mass was concentrated. The over-estimation shown by the reference run in the original manuscript is reduced by several tens of percents. The impact appeared to be quite homogenous spatially and has affected the background level of NO2 to a lesser extend. The replacement of the gas-phase chemical mechanism with the basic version of CBM4 (Gery et al, 1989) has led to more dramatic changes. In this version the high-resolution fields from the TNO inventory were used, and all main peaks are well reproduced again. However, the high background levels were significantly reduced. In general, the pattern better resembled the other models, but not compared to OMI. It points at a shorter lifetime due to chemical transformations in the CBM4 in comparison with the reference SILAM chemistry module.

Summarising, these sensitivity simulations have shown that the modeled background level of NO2 is mainly explained by the specific chemical mechanism as used in SILAM. The level of the column-integrated concentrations is also influenced by other factors,

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such as the intensity of the vertical mixing. A decreased mixing rate has improved the vertical profile of the concentrations. These finding will be briefly discussed in the updated version of the manuscript. We give some special attention to the SILAM model in the revised manuscript, as it appears to give a good match to the seasonal cycle of both OMI and surface observations.

RC3-G: The study further suffers from the fact that both validation data sets, i.e. OMI columns and surface NO2 observations, both suffer from significant uncertainties. The observations therefore only serve as guidelines rather than as validation data sets and no firm conclusions can be drawn regarding good or bad model performance. The authors are not to blame on this but it clearly demonstrates that further efforts are need to enhance the accuracy of the observations. As a consequence, the reader is left with many comparison figures and numbers but doesn't know which one to take as the reference against which the models are to be tested. Unfortunately, only little more can be learned from this analysis than that there are significant differences between models and between models and observations.

AC3-G: We would claim that there are also significant agreements, for instance demonstrated by the reasonable correlations between the datasets. Our work documents the current status of the agreement between models and OMI. We now introduce the ensemble median as a clear reference, see rev#1, AC4-G. In a revised version we removed some of the figures (figure 5-6: individual model results in December 2008 and figure 12: monthly mean average profiles for EU-RAQ region, Italy and the Iberian Peninsula), to condense the presentation of model results.

RC4-G: We do not even learn whether or not the TNO inventory is more realistic for Western Europe than the RETRO inventory, despite the factor of 2.5 difference in emissions. Probably the most interesting result of this study is that whether applying the averaging kernel to the regional models or not did not make a significant difference. However, it would probably be dangerous to generalize this result. The OMI data might have a bias (0-40% in summer) but the relative differences between different regions

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are probably more reliable. Thus, in the comparison of the models with OMI the authors should put more emphasis on relative differences between OMI and models (such as the low values in CHIMERE over eastern Europe).

AC4-G: We acknowledge these uncertainties and the problems to derive general conclusions explaining why models behave differently. Still we think that the reporting of the current state of the RAQ ability to model NO2 is valuable, also for future reference of an updated version of a similar system. Also the differences between the inventories was not reported before in this detail, whereas it is worth to be aware of such discrepancies. We refer to rev#1AC4-5 for more comments on the emission inventory.

AC5-G: Also the reviewer suggests to compare relative differences between OMI and models at different regions. In principle this is a good suggestion, to circumvent possible systematic biases in the retrieval. However, in practice we anticipate problems with such a quantitative analysis, due to differences in local circumstances: e.g. the Iberian Peninsula shows high hotspots at cities with large regions of low background concentrations, the western Europe region has high concentrations on a large scale, but may also be influenced from clean inflow from the ocean. Over eastern Europe the modeled background concentrations seem more related to area-sources. Also biases in the observations are not necessarily identical for different regions. Concerning a more quantitative analysis, we think that the provision of statistics from scatter plots will be more understandable.

Minor points:

RC1: Abstract: The sentence mentioning the small bias when neglecting the averaging kernel is not understandable to the reader in this context. It should first be mentioned more generally what the differences between models and observations are (which is only mentioned later in the abstract) and only then the effect of applying the averaging kernel (or not) may be stated.

AC1: We will reconsider the formulations in abstract, see also rev#1 AC9-G.

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RC2: p22279, line 18: In what form is NOx emitted in the RAQ models?

AC2: This varies fore the different models between 85% (as in BOLCHEM) and 95% NO (EMEP). We will include a comment on this in the description of the RAQ emissions. See also AC5 to rev#1.

RC3- Page 22280, line 10-15: The TNO and RETRO inventories differ strongly over western Europe but are more alike over eastern Europe. So why does this suggest that the RETRO inventory is outdated over eastern Europe (or what do you mean by "these regions")?

AC3: See rev#1, AC4.

RC4:- Line 18: It would be useful to know how much ship emissions add to the total NOx emissions in Europe and thus to know how much the inventories in models with and without ship emissions differ.

AC4: We have provided this information in the revised manuscript in the Table listing the emissions. Ships emissions in the RAQ models are 0.9 Tg N/year (Vestreng, 2003), which contributes of the order of 17% to the total anthropogenic NOx-emissions.

RC5: - Line 21: What about a seasonal cycle in emissions?

AC5: The RAQ models apply different choices in their temporal aggregation, including the seasonal cycle see also rev#1, AC5.

RC6: Page 22282, line 23: What do you mean by "under-representation"?

AC6: With under-representation we refer to the fact that the a-priori profile information is available on a relatively coarse 3x2 degree grid, whereas the measurement pixels are on a much higher resolution. This introduces errors in the retrieval algorithm.

RC7: Page 22286, line 19: Why were emissions "underrepresented by about a factor 2"?

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AC7: This refers to a model error in the implementation of NOx emissions in the MOZART-IFS system: the model only introduced half the NOx emissions. In the revised version we will mention this shortcoming more explicitly, see also rev#1, AC2-G.

RC8:- Page 22288, line 2: I am a bit confused: A few lines earlier it was mentioned that OMI data in June 2009 are much lower than in July 2008.

AC8: See rev#1, AC9

RC9- Page 22289, lines 6-10: I don't think this can be concluded given all uncertainties.

AC9: We agree with the reviewer that the statement is not unquestionable. We have removed it in the revised manuscript.

RC10: - Page 22290, line 11: In meteorology September is not a summer month. So why don't you take August or July rather than September?

AC10: This is a good suggestion. In the revised manuscript we have replaced the figures for September with those from August.

RC11:- Line 16: Increased photolysis is not relevant here. Increased OH is the point.

AC11: The reviewer is correct. We have reformulated this by stating that the photochemical sink from oxidation by OH is larger in summer than in winter.

RC12: - Page 22293, line 3: From a RMS difference you can not infer whether there is a cancellation or not, since the RMS sums up the squared differences (which are always positive).

AC12: We show in figure 10 the RMS of the area-averaged differences between the tropospheric columns with and without application of the AK, which in itself does not indicate a cancellation. But in case of a regional cancelling of differences, Nk and Ntc give similar numbers, but the rms shows a positive number, as we find. So in fact some regional canceling does take place. We also have included an additional figure to show the regional differences with/without application of the averaging kernel.

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RC13: - End of Section 8: Probably a conclusion should be that differences between models are much larger than differences in columns with or without the AK applied.

AC13: This is partly true, as can be seen from many figures, like the comparison of the profiles. Still, we argue that the main conclusions concerning the RAQ models in this section will hold. E.g. almost none of the RAQ models have emissions in the free troposphere, which could suggest that the cancellation of errors with/without AK will take place in general. Also surface concentrations in all RAQ models are significantly lower than the a priori in August 2008.

RC14: - Page 22295, line 26: HNO3 photolysis is very slow (timescale of the order of a week) in comparison to the reaction of NO2 with OH (which limits the lifetime of NOx to only a few hours in summer) and is thus unlikely to make an important contribution to the NO2 column (is HNO3 photolysis not considered in the other models?). The high SILAM columns in summer seem unrealistic and can not be defended in this way.

AC14: The reviewer is correct. We have modified our argumentation, see also rev#2, AC2-G.

RC15: - Figure 9: Why don't you show a similar figure for surface NO2 in comparison to the ground-based observations?

AC15: Our focus in this work is mainly on the analysis of tropospheric NO2 columns. We include a small set of measurements from surface stations to have some independent observations. Although an analysis of diurnal cycle in surface observations is of coarse an interesting suggestion, this would divert from the main theme of this work towards a study of the surface concentrations.

RC16:- Table 4: What do you mean by "spread"? 1 sigma, 2 sigma, full range?

AC16: See also rev#1 AC8.

AC17: all other minor comments from the reviewer are addressed.

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Interactive Comment