Atmos. Chem. Phys. Discuss., 8, S12212–S12219, 2009
www.atmos-chem-phys-discuss.net/8/S12212/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ship emitted NO_2 in the Indian Ocean: comparison of model results with satellite data" *by* K. Franke et al.

K. Franke et al.

Received and published: 19 May 2009

We would like to thank Reviewer 2 for his detailed and helpful comments. Most of the suggestions have been incorporated in the revised paper. In the following we give point-by-point answers to the comments:

General comments:

The authors claim to evaluate ship emissions quantitatively. However, the discussion of errors/uncertainties is quite short. The final message is more or less "it fits quite well", which should be definitely more precise. One important aspect is the fact that the coarse spatial model resolution does not resolve plume chemistry. This is mentioned several times in the manuscript, with according references. However, the possible impact on the results is not discussed quantitatively. In particular, the fact, that good

agreement was observed between satellite and E5/M1 NO2, despite the fact that plume processing has been neglected, is suspicious, and might indicate two (several) effects cancelling each other out. This has to be discussed in more depth.

In response to the reviewer's comments, a more sophisticated error analysis for the satellite data has been included in the revised version of the paper.

The reviewer is correct that the model simulation does not resolve plume chemistry. A parametrisation for sub-grid scale ship plume processes in global models does not yet exist. However, several box model studies on ship plumes showed that the lifetime for NOx is significantly shortened if plume chemistry is considered (e.g. Davis et al., 2001; Song et al., 2003, Charlton-Perez et al., 2009). The shortened NOx lifetime could explain a significant fraction of the overprediction of NOx levels in and near shipping lanes that was found in comparison of global chemistry models with observations (David et al, 2001; Lawrence and Crutzen, 1999). In other words, it is expected that if sub-grid scale ship plume processes are considered in the global model simulation then the simulated enhancement in NOx concentration due to ship emissions would be even smaller which means that the lower ship emission estimate would agree even less with the satellite observations. We have made this clear in the revised manuscript.

The obvious shift of the shiptrack between AMVER and E5/M1 is mentioned and explained by the fact that the ship track is close to a model grid latitudinal boundary. However, since the AMVER pattern was used for ship emissions in the model (16003 3-4), the model maximum should be found according to the AMVER maximum. The latter is found at approx. 6°N throughout the year (Fig. 3c), which is definitely north of the maximum E5/M1 grid box (2.8°N-5.6°N). From the AMVER emissions, I thus would expect the model peak in the next latitudinal band northwards (5.6°-8.4°N).

The shift in the location of the shipping lane between ship activity data and the model is the result of a two step regridding process. First the ship activity is regridded from a 0.1 degree grid to a 1 degree grid. In this step the majority of the ship traffic is allocated

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to the box centered at 5.5 degree north. This is regridded in the second step into the box from 2.8 to 5.6 north. This information has been added to the revised manuscript.

For the comparisons performed here, the exact position of the shipping lane in the emission data is not relevant as the values were integrated accordingly.

The authors use GOME, SCIAMACHY, and GOME-2, but ignore OMI data. Since OMI observations take place at a quite different local time (2 p.m.), they might in particular provide valuable insights on the aspect of diurnal variation of NO2.

We agree with the reviewer that OMI data are a very interesting additional source of NO2 data for shipping emissions, in particular as they are taken at a different time of day. However, while for the other three satellites instruments we use a fully consistent retrieval reducing possible instrument biases, for OMI we have to rely on slant columns provided by the operational NASA product. Therefore, any differences observed can be either from real changes in atmospheric NO2 content or from subtle differences in the spectral retrieval (choice of fitting window, cross-sections, polynomial etc.). Considering these uncertainties, we decided to mainly stick to SCIAMACHY data in the manuscript but to include OMI data in the discussion of the temporal evolution (Fig. 6).

16000 9-11: How do you judge about the overestimation of ship NOx in models due to model resolution? What is the impact of this effect on your study (see also below)?

See response above.

16002 11-12: Giving a relative number for the accuracy is probably appropriate for polluted regions with high NO2 columns (where the uncertainty is mainly due to the air mass factor), but are misleading for low NO2 levels: In the extreme case of a column of 0, this value would be free of error! So over "clean" regions, additive errors are quite important, as may arise from the stratospheric estimation or from unidentified spectral structures that may be interpreted as NO2 in the fitting process. These systematic biases probably partly cancel out by considering the differences of neighboring

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regions, but still it cannot be excluded that the derived difference in mean columns has a systematic bias.

We agree with the reviewer that the error discussion for the satellite derived NO2 columns was too short in the original manuscript. We therefore have added a more detailed discussion of error sources and the additive and multiplicative components of the overall uncertainty.

16003 3-4: If AMVER is used for the spatial distribution of emissions, then why is the E5/M1 shiptrack shifted one grid box further south??? (see also 16005 7-8)

As explained above, a two step regridding process has been applied to the inventory data which leads to this shift in apparent emission location. For the conclusions of the paper, this shift should not be relevant.

3.2 The authors discuss differences of GOME, SCIAMACHY, and GOME2 results, giving 3 possible explanations. However, I would expect that there are also systematic differences between the 3 NO2 products from the different sensors. The authors might discuss this aspect first, making use of the temporal overlap between GOME and SCIAMACHY as well as between SCIAMACHY and GOME2. In addition, OMI data should be compared to SCIAMACHY and GOME2 for consistent time periods. Afterwards, the authors can go through (i)-(iii), in which the addition of OMI data helps in particular to judge about diurnal variations.

We agree with the reviewer that the uncertainties of the three data sets contribute to the observed differences. As mentioned above, a more detailed error discussion has been included in the revised manuscript addressing this point.

The second suggestion to use the overlapping parts of the time series to quantify instrumental biases is a good suggestion, and in fact in Fig. 6 of the original manuscript, this comparison is shown. However, as the measurements are taken at different times of the day and with different sampling, perfect agreement can not be expected even

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in this comparison, and the contributions from time of day variations and instrumental biases can not be disentangled.

With respect to the use of OMI data, we followed the advice of the referee although the OMI data product is not as consistent with our other NO2 columns as we would wish (see discussion above). Therefore, this part of the comparison in Fig. 6 has to be taken with care.

3.3 This section has to be more quantitative.

- What is the expected effect of in-plume chemistry? For this particular ship track, I expect quite special conditions: due to the narrow track with high ship frequency, average NOx (and probably VOC) levels are far above natural background, and, on top of this, there are point-source-like ship emissions. This is different from the single-ship study in Franke et al., 2008.

- If the agreement is good, despite neglecting model resolution effects, could this indicate two (or more) compensating effects?

- What about aerosols? What is their impact on the satellites sensitivity (air mass factors) and on the NOx lifetime?

In the end, the authors should give an estimate of ship emissions from their comparison study including errors.

Regarding the effect of ship plume chemistry, please see our response above.

Aerosols can have a significant impact on the radiative transfer in the atmosphere. For satellite nadir observations of tropospheric absorption, both enhancement and reduction of sensitivity can result from aerosols, depending on their vertical distribution relative to the NO2 and their single scattering albedo. Sensitivity studies assuming different amounts of aerosols with different SSA have shown, that for an aerosol that is mixed within the NO2 layer, the net effect is surprisingly small (of the order of 5 % for AOD 0.1 and of the order of 30% for AOD of 0.5 for a well mixed layer of 700m) unless

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the aerosol is highly absorbing. We agree with the reviewer that aerosols have the potential to interfere with the retrieval, and it would be very interesting to investigate the effect using airborne measurements of shipping plumes. For the current study, we have to limit ourselves to using the results of the sensitivity studies for the error assessment.

From the S-B2 comparison in Fig. 8 (that should be less affected by biomass burning etc.), there are several points that are closer to the dotted line that to the straight line, and from the given error bars one cannot refuse 1:1 nor 2:1. In so far, the statement in the abstract that "the results do not support ... 3-4 Tg" is not supported by the presented data.

In the revised paper, we have replaced this comparison to a direct comparison between satellite and model data using two different assumptions for the shipping emissions. The conclusions from this comparison are very clear showing that only the larger emission estimates lead to model values which agree with the satellite data within uncertainties. The corresponding sections have been reformulated accordingly.

From the error bars shown in Fig. 8, and, in addition, uncertainties arising from inplume chemistry, aerosols, and other systematic errors, I expect a resulting uncertainty of at least 50%.

4 The conclusions should be updated depending on the extended error discussion and new results from OMI data.

We have updated the conclusions reflecting the results from the extended error discussion and the additional data.

Minor comments (Line numbers referring to discussion paper):

15998 3-5: I suggest to mention the satellite instruments chronologically, i.e. start with GOME.

As the main focus is on SCIAMACHY data we decided to keep the order of listing the instruments.

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15999 20-22: I couldn8217;t find that number (56 ppbv), neither by text search nor from any figure, in Eyring et al., 2007.

Thank you for pointing out this typo - it should read 5-6 ppbv and is corrected in the revised paper.

16001 13: If you mention the high-res mode of GOME, you should also mention that it is applied only every tenth day, and, especially, it is gained at the cost of swath width, i.e. global cover of the high-res mode is quite sparse. But since this is quite irrelevant for this study, I suggest to skip it and just mention 40x320 as "nominal" GOME resolution (SCIAMACHY also has different observation modes with different spatial resolutions).

The Hi-res mode of GOME skipped in revised version as proposed by the reviewer.

16001 25-26: You should clarify that there are many possible definitions for a reference sector, and that 180° E-220° E is your actual choice. I suggest to change the description to 180° W - 140° W, in accordance with the ticks in Fig. 1.

The description of the reference sector has been changed to point out that the region 180 - 140 W is our actual choice.

16006 9: ... and model

Section heading was clarified.

16009 the aspect (iii) should be discussed in a separate paragraph, as (i) and (ii), before summing up (i)-(iii).

Aspect (iii) is now discussed in a separate paragraph as suggested by reviewer.

16020 The authors might rethink the choice of colour bar and -range; in particular in Fig. 3b, only 3 different levels of NO2 can be recognized.

The specific color scale for the NO2 TEC was chosen in order to a) be the same in every figure and b) not to draw the attention to detail far below errorbars. Therefore we

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decided to keep our scale as it was in the discussion paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 15997, 2008.