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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Constraints on ship NO_x emissions in Europe using GEOS-Chem and OMI satellite NO_2 observations" *by* G. C. M. Vinken et al.

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We thank reviewer #2 for reviewing our paper and the provided comments and detailed specific comments. Please find a detailed discussion on the comments below. We adapted our manuscript in line with these recommendations. We marked updates in our manuscript with a blue text color.

Major Comments

1) Consideration of emissions during the 2.5 h time step in the plume-in-grid approach (P 19357): The authors correctly point out that the satellite view is not that of a plume

of age 2.5 hours but rather a composite of plumes which one could assume to be formed continuously in the shipping lanes. Why this can be modelled by simply adding fractions of NO_2 *left after 2, 1, and 0.25 h is not clear to me at all. Please explain. It would also be good to comment on the effect the continuous emissions have on the background* NO_2 *levels mixed into the plume in the plume model.*

To account for the emissions in the 2.5 hours before the satellite observation, we discretize these emissions in 3 parts, and then add the overall effect to the standard model output, as illustrated in Fig. 1. We multiply the emissions that were released between 1.5 and 2.5 hours ago (1 hour of emissions) with the fraction of NO_x remaining computed with the plume model after 2 hours. For the second part we multiply the emissions between 0.5 and 1.5 hours ago with the fraction of NO_x remaining after 1 hour. The last part consists of the emissions between 0 and 0.5 hours ago, and is multiplied by the fraction of NO_x remaining after 15 minutes. We have now clarified this in the manuscript. We take into account background NO_2 levels in our plume-in-grid approach (it is a parameter in the look-up table), hence the (reduced) ship emissions are superimposed on the enhanced background concentrations. The effect of these higher background NO_2 concentrations is a higher fraction of NO_x remaining (see e.g. Fig. 2 in Vinken et al. (2011)).

2) Comparison of GEOS-chem and OMI data (Sec. 2.3): here, the average over the full European domain is compared, showing quite reasonable agreement. However, for the present study it would be much more relevant to compare the NO_2 *values in the regions with ship emissions and in polluted regions close to these areas. Judging from Fig. 3, this will give quite different results (higher model than measurement* NO_2 *in the Mediterranean, much higher OMI than GEOS-chem* NO_2 *in the North Sea). Please extend Fig. 4 with additional regions.*

We agree that for this study it is most important to compare OMI and GEOS-Chem over regions with ship emissions. This is exactly why we did a detailed comparison for ship tracks in several seas in Fig. 7 and Fig. 9 (now Fig. 8 and Fig. 10) and Table 2. These

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comparisons show different results than the comparison in Fig. 3 and Fig. 4 (indeed higher modeled than observed NO_2 in the Mediterranean and the North Sea, and lower modeled columns than OMI observed in the Baltic Sea and the Bay of Biscay). The main motivation for showing Fig. 3 and Fig. 4 is that GEOS-Chem European nested-grid has never been evaluated, and by comparing model and observations for this region the results can be compared with previous model evaluations by Huijnen et al. (2007) (Fig. 6 for region mid/south-RAQ) to obtain a sense of model skill. They found (for 2008/2009) that their ensemble median is on average 50% below OMI observations in summer, and only has a small bias in winter. Our GEOS-Chem simulations show a stronger correlation and close match in summer, providing confidence in the ability of the GEOS-Chem nested grid model to simulate tropospheric NO_2 columns over Europe. We have now included this discussion in Sec. 2.3.

3) The same argument holds for the comparison of DOMINO2 and DOMINO2GC – the interesting changes are over the shipping lanes and there I would be surprised to see differences of only 10% as stated in Sec. 3.3 – I expect better spatial resolution + plume chemistry to result in larger differences over shipping lanes also to increasing instead of decreasing values. Please comment.

Indeed changes over shipping lanes can be stronger than the 10% (between DOMINO2 and DOMINO2_GC) stated in Sec. 3.3. In addition to the better spatial resolution and plume chemistry this is also driven by the different emissions in GEOS-Chem compared to TM4 (as discussed in Sec. 2.3.). We have now included a difference plot between DOMINO2 and DOMINO2_GC for 2005 over the European domain in our revised manuscript (Fig. 5). Different emissions in GEOS-Chem (EMEP vs. POET 97) can be observed by the decreases of tropospheric NO2 over western Europe where EMEP emissions are lower, and by increased NO_2 columns over eastern Europe (here EMEP emissions are larger). The effect of the higher resolution can be observed by the increase in emissions in Spain (Barcelona and Madrid); a higher resolution results in more localized emissions (and hence a higher simulated column $->$ lower AMF

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$->$ higher observed column). For the Mediterranean Sea NO_2 columns change by +20 to +45%, for the Bay of Biscay and the North Sea by -10 to -20%, and for the Baltic Sea by -20 to 30%.

4) Selection of unpolluted scenes (Sec. 2.4): This is not explained at all! Which criteria have been applied to select the data used in the inversion? Please elaborate.

We filtered for at least 90% of the area having a cloud radiance fraction < 0.5, and not a single negative NO_2 column. Furthermore, to filter for outflow, we required the NO_2 values in background pixels (adjacent to the ship tracks) to be lower than 2×10^{15} molecules cm^{-2} for the North Sea, the Baltic Sea, and the Mediterranean Sea, and $< 1.5 \times 10^{15}$ molecules cm^{-2} for the Bay of Biscay. We included this in Sec. 2.4 and the caption of Fig. 5 (now Fig. 6).

5) It is not entirely clear to me how Eq. 4 is being used – is that an iterative process? If so, does it converge? Please comment.

We have included additional steps in the description of Eq. 4 in our manuscript, and also indicate the different simulations we have done. Indeed this approach converges, as shown by the close match in Fig. 9 (now Fig. 10).

6) Displacement of emissions in Bay of Biscay: How can that be the case (I thought that the emission inventory used is based on actual ship positions) and how has the inversion corrected the misplacement (or was this done manually in an ad hoc way)? Please explain.

As a priori emission inventory we use the EMEP emissions totals combined with the location of the AMVER-ICOADS inventory for the Mediterranean Sea. For all seas other than the Mediterranean Sea, this a priori emissions inventory is identical to the EMEP inventory. This EMEP inventory is based on the distance each ship covers between ports (from the Lloyd's Register of Shipping (Vestreng et al., 2003)). As was show for the Mediterranean Sea, this does not always result in ship emissions at the

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correct position. Our inversion does not correct the location of the emissions, so prior to the inversion we shifted the emissions in the (combined) inventory to match the OMI location. We included this in Sec. 3.4.

7) Top down emissions: In the paper it is claimed repeatedly that 39% of all ship emissions are constrained by the inversion. However, looking at the coverage of OMI data used, I can't believe that this is the case. I rather assume that the 4 scaling factors derived from analysis of Figs. 7 have been applied to the much longer tracks shown in Fig. 8 assuming that they are representative for the full shipping lane. While this is probably a reasonable assumption, I feel that it must be clearly stated in the paper and would recommend not to put so much emphasis on the 39% which are not really the fraction of emissions directly constrained by observations.

Indeed, we derived 4 scaling factors from our constraints, and applied them to the ship tracks shown in Fig. 8d (now Fig. 9d). NO_x emissions in these ship tracks correspond to 39% of all European ship NO_x emissions. To clarify this in the manuscript we now indicate the areas of the ship tracks where we derived these constraints in Fig. 5 and Fig. 8 (now Fig. 6 and Fig. 9), and also included this in Sec. 3.5 (and in the abstract).

8) Along ship track averages: How have these been computed? Considering the model resolution of 0.5 x 0.67 degrees, I do not see how you can have so smooth curves in Figs 7 and 9. How was the integral taken – have the boxes in Fig. 5 been rotated, interpolated, and integrated along the line visible in the emission inventories? Where did the integration start and stop? In particular in the Baltic Sea where quite some good will is needed to discern a shipping lane in the data it is important to explain exactly what was done to create the data which are the basis of the inversion.

The boxes were indeed rotated (and interpolated), and averaged along the ship track. We now include the area over which the integration has been performed in Fig. 5 and Fig. 8 (now Fig. 6 and Fig. 9). We have also extended our description of this integration in Sec. 3.4.

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9) Please also explain how exactly the linear background was found and why the model data were treated this way instead of just using two runs, one with and one without shipping emissions.

To illustrate our background correction we now include Fig. S3 in the Supplementary Material to show a cross-section without the background correction, and indicate how the linear background was fitted. Due to considerable non-linearities in the in-plume NO_x chemistry, a run without shipping emissions is unlikely to properly represent the contribution of ship emissions to the background. The non-linearity in the ship tracks is illustrated by the β values being unequal to 1, illustrating that changes in NO_x emissions do not linearly follow changes in columns. Furthermore such a method could not be applied to the OMI observations, and would lead to inconsistencies in the comparison. By using this background correction method we ensure that OMI and GEOS-Chem have the same correction.

Minor Comments

Introduction: I'm not an expert but to my understanding, proposed legislation does not set limits on nitrogen oxide emissions but rather on technology used in new ships.

We changed this in the revised manuscript.

P19358, L27: an moderate => a moderate

We changed this in the revised manuscript.

P19359, L24: OMI retrieves => The first step of retrievals on OMI data yields

We changed this in the revised manuscript.

P19365, l10: ... OMI NO_2 change from a priori changes is never larger...

We changed this in the revised manuscript.

P19366, L3: base => based

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We changed this in the revised manuscript.

P19366, L12: emissions => emissions in GEOS-Chem

We changed this in the revised manuscript.

P19366, L25: Is that differences between columns or between enhancements relative to a background?

This is difference between enhancements relative to background. We adapted this in the manuscript.

Fig. 8: Is that for 2005?

Indeed, this is for 2005. We updated this in the manuscript.

Refences

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