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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 #1 for the review and constructive comments provided. Please find our detailed replies to the comments below. We adapted our manuscript in line with these recommendations. We marked updates in our manuscript corresponding with this review with a red text color.

Comments

1) Abstract: The abstract is quite long, and I recommend to shorten it. However, I miss a sentence on the idea how the constrained emissions are derived.

We shortened the abstract as indicated in the revised manuscript. Furthermore we added an additional sentence on how the emissions are constrained.

2) P19352 L4: Why was the time period restricted to only two years, where OMI provides a much longer time period?

Indeed, OMI NO_2 observations are available over a longer time-span (2005-now), but it was out of the scope of this research to study all years. This study serves as a proof-of-concept, and future research could focus on how the emissions change over the OMI time period.

3) P29352 L21: Please reword. The satellite "observation" is not affected by a model, but the derived vertical column is.

We adapted this in the manuscript (in the conclusions section, as this line was removed from the abstract).

4) P19355 L18: I miss a clear definition of the investigated areas. I propose to put a figure here, similar to Fig. 6a, where the current knowlegde on shipping routes is reflected, and the investigated regions are marked by a box. In addition, the region boundaries should be added to table 2.

We included more information on the ship track boundaries in Table 2, and indicated the investigated areas in Fig. 5 (now Fig. 6) and Fig. 8 (now Fig. 9), and the caption of Fig. 7 (now Fig. 8).

5) Section 2.1: I haven't fully understood the concept of the combined CTM/plume model: In Vinken et al., 2011, a box model is used to study individual ship plumes. But how are these results merged into the CTM? Is each individual ship represented by a ship plume? Or, as I assume, are the results from the plume model scaled into the CTM? How is this scaling done, by number of ships or by average emissions? How could this effect your study?

The integration of the plume model simulations in the GEOS-Chem CTM has been

described extensively in Vinken et al. (2011). For completeness, we use a Gaussian plume model to simulate the non-linear chemistry in expanding ship plumes in the first 5 hours. We store the results of the plume model in a look-up table, which provides the fraction of NO_x remaining and integrated Net Ozone Production Efficience (NOPE) as a function of 7 environmental parameters. For every grid cell with emissions in GEOS-Chem, we look-up the fraction of NO_x remaining, and multiply the emissions in this grid cell with this value (scaling). In this approach we assume that the emissions in the GEOS-Chem grid cell consist of a collection of plumes with an average emission strength. The effect of this assumption is minor, as we found in Vinken et al. (2011) that the fraction of NO_x remaining does not depend strongly on the emission strength (also see Fig. 24 in Vinken (2010)). This method might introduce additional errors, as discussed in Vinken et al. (2011), and we include these errors in our final error estimates (Sec. 3.6). We have extended our description of this method in our manuscript (Sec. 2.1).

6) P19358 L8/L15: It took some time until I understood this paragraph. Please avoid switching the order of what is higher/lower compared to what.

We partially rewrote this paragraph to improve readability.

7) P19359 L16: "dominates ... over oceans": Please comment on this with respect to this study: It might be different over ship tracks!

Indeed, this is different over ship tracks, as these represent more polluted scenes. These errors are over clean ocean air, we clarified this in the manuscript.

8) P19360 L13: How sensitive are the results on clouds? Information on this can be gained by varying the cloud radiance fraction threshold.

In addition to the cloud radiance fraction threshold of 0.5 for individual pixels, we also filter for all pixels in the entire ship track area to have a cloud radiance fraction threshold < 0.5. This is a tight filter for clouds, so changing these values isn't expected to lead

to strong differences in the results. A tighter cloud radiance fraction filter will lead to less data, and loosening the filter will result in more clouded pixels to be included in the average. We use a value of 0.5 for the cloud radiance fraction threshold (corresponding to cloud fraction approximately $<$20%), as NO_2 retrievals retained with this filter show best agreement with surface observations (see Fig. 1 in Geddes et al. (2012), and Fig. 7a in Schaub et al. (2006)).

9) P19361 L12: Why should lower emissions result in higher AMFs? Please explain!

Lower emissions lead to lower concentrations of NO_2 in the atmosphere (see profiles in Supplementary Material Fig. S1), resulting in a decreased absorbance of NO_2 in these layers (and an on average increased light path through the atmosphere). This is accounted for by a higher AMF (see Fig. S2), and hence lower tropospheric vertical NO_2 column (Martin et al., 2002; Eskes and Boersma, 2003; Boersma et al., 2004). Barkley et al. (2012) also showed that lower emissions lead to higher AMFs in the retrieval of HCHO vertical columns. We now also include Fig. 5 in our manuscript, indicating the effect of higher AMFs on the tropospheric NO_2 columns over Europe, and include more discussion in Sec. 2.3.

10) Section 2.4: How is the selection done in detail? Are there some thresholds applied, or was it done by visual inspection only?

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).

11) P19362 L2: Fig. 3 shows a map of OMI NO_2, in which some ship tracks can be recognized or guessed. In my print out, the mediterranean ship track is much better visible in Fig. 6b than in Fig. 5c. Why? Please optimize Fig. 5.

In Fig. 5c (now Fig. 6c), we have applied our filtering as described in section 2.4. Fig. 6b (now Fig. 7b) includes all (valid) OMI NO_2 observations, and therefore has a different sampling. We chose a different scale in Fig. 5c (now Fig. 6c) than in Fig 6b (now Fig. 7b) because this better shows the ship tracks in the Bay of Biscay.

12) Section 3.2: How is beta calculated? Is it the spatio-temporal mean (complete box, complete month/season) for Delta N and N?

Beta is calculated from the along ship track averages (see Fig. 7, now Fig. 8) for the different simulations. For the Baltic Sea, the North Sea and the Bay of Biscay these are calculated from the cloud-free days of 2005 that meet our filter criteria (see Sec. 2.4), and for the Mediterranean Sea from seasonal averages filtered by these criteria. We added this information to Sec. 3.2.

13) Equation 4: I can not comprehend this equation:
- If there is some kind of iterative approach, the terms N_OMI-N_GC should be different in the both brackets.
- I do not understand why there is just a second additive term multiplied by gamma: gamma affects N_OMI, so I would expect to have it as factor at N_OMI, not the whole term!?
- Please provide the maths leading to Eq. 4: A change of emissions causes a specific change of N_GC, and, due to gamma, also of N_OMI. I tried to write down delta E as function of N_OMI and N_GC, using Eqs. 1 and 2, but found it to be not trivial to relate this to (N_OMI-N_GC)/N_GC, as in Eq. 4. So please justify Eq. 4 in detail and show how this equation follows from Eqs. 1 and 2, and clearly state what assumptions have to be made.

We have improved the description of Eq. 4 in the manuscript. Indeed it is an iterative approach, and we have included subscripts in the equation. We ran four simulations with the GEOS-Chem model:
Simulation 1: Standard model run (emissions at 100%)

Simulation 2: Run to allow calculation of β and γ; Ship emissions perturbed by relative difference in observed and simulated columns; factor $((N_{OMI,1} - N_{GC,1})/N_{GC,1})$
Simulation 3: Run with constraints applied and accounting for non-linearities, with a priori emissions increased by $((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot (\beta_1 + \gamma_1 \cdot \beta_1)$
Simulation 4: Additional iteration of constraints, to account for the strong changes, top down emissions of simulation 3 are increased by $((N_{OMI,3} - N_{GC,3})/N_{GC,3}) \cdot (\beta_2 + \gamma_2 \cdot \beta_2)$

From the first run we determine the relative difference between OMI observed and GEOS-Chem simulated NO_2 columns, $((N_{OMI,1} - N_{GC,1})/N_{GC,1})$.

Then we run GEOS-Chem a second time, with ship emissions perturbed by $((N_{OMI,1} - N_{GC,1})/N_{GC,1})$. We use simulations 1 & 2 to calculate β_1 and γ_1:

$$\begin{aligned} \beta_1 &= \frac{\Delta E/E}{\Delta N_{GC}/N_{GC,1}} \\ &= \frac{(N_{OMI,1} - N_{GC,1})/N_{GC,1}}{(N_{GC,2} - N_{GC,1})/N_{GC,1}} \end{aligned}$$

and

$\gamma_1 = \frac{(N_{OMI,2} - N_{OMI,1})/N_{OMI,1}}{(N_{GC,2} - N_{GC,1})/N_{GC,1}}$

The conventional approach (see Lamsal et al., 2011) would be to update the a priori emissions inventory by:

$E_{top\,down} = E_{a\,priori} + \left(\frac{N_{OMI,1} - N_{GC,1}}{N_{GC,1}}\right) \cdot \beta_1 \cdot E_{a\,priori}$

as this accounts for the non-linear response of NO_2 columns to changes in NO_x emissions.

However, we know that also the retrievals will change because of changes in the a priori NO_2 profile, i.e. by a factor $((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot \gamma_1$. To account for this

(in first order), we increase the a priori emissions by an additional factor $((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot \gamma_1 \cdot \beta_1$ (to increase the GEOS-Chem column by the same factor as the expected increase in OMI column):

$$E_{top\,down,1} = E_{a\,priori} + \left(\frac{N_{OMI,1}-N_{GC,1}}{N_{GC,1}}\right) \cdot \beta_1 \cdot E_{a\,priori} + \left(\frac{N_{OMI,1}-N_{GC,1}}{N_{GC,1}}\right) \cdot \gamma_1 \cdot \beta_1 \cdot E_{a\,priori}$$

Although this additional increase of the a priori will again lead to a change in OMI we neglect these higher order terms.

We found that for the Baltic Sea and some seasons in the Mediterranean Sea emission changes were large and an additional iteration was needed. For this we calculated new β and γ factors from Simulation 2 and Simulation 3;

$$\begin{aligned}
\beta_1 &= \frac{\Delta E/E}{\Delta N_{GC}/N_{GC,2}} \\
&= \frac{1}{(N_{GC,3} - N_{GC,2})/N_{GC,2}} \cdot \\
&\quad \left(\frac{E_{a\,priori} + ((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot (\beta_1 + \gamma_1 \cdot \beta_1) \cdot E_{a\,priori}}{E_{a\,priori} + (N_{OMI,1} - N_{GC,1})/N_{GC,1} \cdot E_{a\,priori}} - \right. \\
&\quad \left. \frac{E_{a\,priori} + ((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot E_{a\,priori}}{E_{a\,priori} + ((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot E_{a\,priori}} \right) \\
&= \frac{1}{(N_{GC,3} - N_{GC,2})/N_{GC,2}} \cdot \\
&\quad \left(\frac{((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot (\beta_1 + \gamma_1 \cdot \beta_1) - ((N_{OMI,1} - N_{GC,1})/N_{GC,1})}{1 + ((N_{OMI,1} - N_{GC,1})/N_{GC,1})} \right) \\
&= \frac{((N_{OMI,1} - N_{GC,1})/N_{GC,1}) \cdot (\beta_1 + \gamma_1 \cdot \beta_1 - 1)/(1 + ((N_{OMI,1} - N_{GC,1})/N_{GC,1}))}{(N_{GC,3} - N_{GC,2})/N_{GC,2}}
\end{aligned}$$

and

$$\gamma_2 = \frac{(N_{OMI,3}-N_{OMI,2})/N_{OMI,2}}{(N_{GC,3}-N_{GC,2})/N_{GC,2}}$$

Then the final constraint on the emissions is:

$$E_{top\,down,2} = E_{top\,down,1} + \left(\frac{N_{OMI,3}-N_{GC,3}}{N_{GC,3}}\right) \cdot \beta_2 \cdot E_{top\,down,1} + \left(\frac{N_{OMI,3}-N_{GC,3}}{N_{GC,3}}\right) \cdot \gamma_2 \cdot \beta_2 \cdot E_{top\,down,1}$$

Simulations with this final run result in the cross-sections of Fig. 9 (now Fig. 10).

14) Section 3.5/Fig. 9: I am surprised and also worried by the strong change in OMI compared to Fig. 7. As I am not convinced by the correctness of Eq. 4, I suspect that something might have went wrong here. What would happen if a further iteration would be applied? Would this further decrease the OMI columns in the Mediterranean?

The strong change in OMI NO_2 retrievals may be expected as a result of the strong change in emissions, and hence the NO_2 profiles and resulting lower AMFs (as was also found earlier in Barkley et al. (2012) for HCHO vertical columns). Although the emission changes are very large, the final top-down inventory is in close agreement with the AMVER-ICOADS inventory for the Mediterranean Sea. As the relative differences between the model and observations are only small in Fig. 9 (now Fig. 10) (<10%), an additional iteration would only result in a small change in the columns.

15) I would also like to ask for more information on what's going on here; so please provide figures/tables on the GEOSchem profiles and AMFs for the shiptracks for initial and constrained emissions (e.g. as supplementary material).

We have now included an additional figure (Fig. S1) showing the GEOS-Chem profile for June 2006 within the main ship track of the Mediterranean Sea (simulated with a priori emissions), and the GEOS-Chem profile for the simulation using the top-down constrained emissions. For this location, the NO_2 tropospheric vertical column density decreases strongly (almost by a factor 5) in the lowest model layer. This is also

reflected in the differences between the AMFs (Fig. S2); higher AMFs in the Mediterranean Sea (+20%) and the North Sea (+10%), and lower AMFs in the Bay of Biscay (-25%) and the Baltic Sea (-20%).

16) I am also missing some discussion on why beta and gamma are that different for the different ship tracks.

Differences in β and γ are driven by the magnitude of emission changes and local chemical regime. For example, in the Baltic Sea we impose strong emissions changes, and the resulting β's are small (comparable to a reduction in β from $\sim$2 to $\sim$0.7 found in Lu and Streets (2012) for increasing emissions of power plants). Emissions changes for the North Sea and the Mediterranean Sea are similar, however the calculated β values ($\sim$0.6 for North Sea, $\sim$0.8-0.9 for the Mediterranean Sea) indicate different chemical regimes. This is consistent with findings shown in Fig. S1 in Lamsal et al. (2011), which indicates that β values are lower for wintertime and polluted areas, and higher β values correspond with summer and less polluted areas. γ values also depend on environmental conditions, i.e. smaller for cleaner background areas (the Baltic Sea and the Bay of Biscay). We have included this discussion in Sec. 3.2.

17) Table 2: Please add a column on the a-priori input emissions (the combined EMEP/AMVER emissions used in GEOSchem).

The combined EMEP/AMVER inventory has the same emission totals as the EMEP inventory (the AMVER-ICOADS locations for the Mediterranean Sea were used in the combined inventory, scaled to the EMEP total). We have added this to the caption of the table.

18) Figs. 6/8: Please indicate the areas defined as ship track as boxes in all maps for orientation.

We have included the areas used in the constraint in Fig. 5 and Fig. 8 (now Fig. 6 and Fig. 9).

19) Fig. 7: Add the a-priori emissions, as in Fig. 9.

We have included the a-priori emissions in Fig. 7 (now Fig. 8).

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