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Atmospheric Chemistry and Physics Discussions

Interactive comment on "What can we learn about ship emission inventories from measurements of air pollutants over the Mediterranean Sea?" *by* E. Marmer et al.

E. Marmer

elinamar@gmx.net

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Answer to Reviewer 1

Thank you for your detailed review and constructive comments which helped us to improve our manuscript. We have revised it following your recommendations, please find our detailed replies below:

1) Page 7156, line 1: Where is the two order of magnitude difference? Not recently observed in large-scale global models, even across regions around ports or heavy traffic regions. Not among port-based inventories, as far as I know. This appears to be generalized from a specific pollutant (NMVOCs), and may be misleading to readers.

Reply: We agree that the two orders of magnitude are only true for NMVOC emissions and are not typical for other compounds.

However, globally we find a difference between the minimum and the maximum estimate of a factor of 18 for CO, 10 for BC and 4.4 for NOx, while OC and SOx differ by a factor of 2. For NMVOC, the difference is over 200 times between Eyring et al.(2005) and EDGAR FT2000, but even NMVOC emissions in the new version of EDGARv4 are a factor of 10 higher than reported by Eyring et al. So there definitely is an improvement, but the disagreement is still very large. In the Mediterranean Sea, when regional inventories are considered, we find a factor of 42 for CO, 15 for BC, 4.4 for NOx, 14 for OC, and 4 for SOx. Looking at these important differences we can conclude that the inventories are converging but have not yet converged.

The sentence has been therefore changed to "... in a difference ranging from a factor of 1.5 to even an order of magnitude".

2) Page 7156, line 6: This statement is overstated at least. Evaluating the validity and consistency of ship inventories has been a primary focus of many, and this work implements a 2003 New Directions (doi:10.1016/j.atmosenv.2003.08.003) call to "design experiments that pursue closure and enable critical uncertainties in the linkages between emissions and atmospheric impacts to be reduced." The work to develop network models for shipping published by Wang et al, ES&T, 2007 demonstrated such evaluations as did the later comparison cited in this paper as Wang et al, 2008); the older work evaluating the calculation system for Finnish waterborne traffic emissions (MEERI, http://lipasto.vtt.fi/meerie/index.htm) also worked to do this, although without field observations as done in this work; the very recent work published by Lack et al, JGR-Atmospheres, 2009 also demonstrated this with a strong emphasis on field observations.

Reply: We agree that a number of earlier studies have been devoted to the evaluation of ship emission inventories. Numerous studies made intercomparisons between

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different emission inventories or used stack measurement to verify emission factors. We have now included references to those studies of direct relevance to our work, i.e. those studies which compared ambient atmospheric concentrations to verify emission inventories. The sentence “little has been done ...” has been canceled from the Abstract, references to other comparisons with observations have been added in the Introduction: “Capaldo et al., (1999), Davis et al., (2001), Lawrence and Crutzen, (1999) and Kasibhatla et al., (2000) have included ship emissions of SO2 and/or NOx into chemistry transport models to evaluate them with aircraft measurements over Pacific and North Atlantic. While including SO2 shipping emissions have improved the model performance with respect to the observations, ship emissions of NOx seemed to be overestimated. Beirle et al. (2004) have quantified shipping emissions of NOx over the shipping route connecting Sri Lanka to Indonesia using data from GOME remote sensing instrument. Data from SCIAMACY, a remote sensing instrument with finer resolution, was used to verify ship emission estimates over the Red Sea by Richter et al. (2004). Both studies were in agreement with existing inventories for the respective regions. Franke et al. (2008) combined data from both instruments to verify all published NO2 emission estimates from ships in the Indian Ocean and found the best agreement with the highest emission estimates“.

3) Page 7156, lines 19-21: So densely populated regions, i.e., regions with significant industrialization and/or human activity, contribute relatively more pollution than ships in such regions, even as they attract heavy ship traffic activity.

Reply: Sentence added: “... because densely populated regions with significant human activity contribute relatively more to air pollution than ships, even if these regions attract a lot of ship traffic.”

4) Page 7156, lines 22-27: And additional work is needed to improve the correlation among inventories and observations. This appears to be worthwhile, where EMEP has provided such work and produces the best match. This work is recommended as the next best way to reduce uncertainties further.

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Reply: These recommendations are given in Conclusions. Our conclusion is rather, that although obviously more work needs to be done in this direction, we do not expect to substantially reduce the uncertainties in emission inventories in the near future and rather should consider the whole range of possible ship emission estimates when producing scenarios and projections.

5) Page 7157, line 22: Sentence reading, " The revised Annex VI will enter into force in July 2010" mistakenly implies to the general audience that the measures required by Annex VI will be implemented in 2010. I know the authors understand the difference, namely that the measures will be phased in over the decade following, but it could be made clearer.

Relpy: Sentence changed: "... the revised Annex IV will enter into force in July 2010, the emission reduction measures will be phased in over the following decade".

6) Page 7159, lines 4-6: While clear, this may not be the best speciation. Please discuss with regard to the Lack et al, JGR, 2009 article at least. Where is the sulfur particle component? Or are you only referring to primary PM (so called)?

Reply: Yes, we are referring to primary PM. The primary emitted sulfur is considered in the SOx (SO2 + SO4) emissions, 2.5% of SOx is emitted as sulfur. Sentence added:

"This speciation varies across the literature, from 11% OC and 5% BC (Eyring et al., 2005) over 30% OC and 15% BC (Lack et al., 2009) to 49% OM and 6% BC (Wang et al.. 2008) ."

7) Page 7160, lines 7-10: Nicely stated. Thank you.

8) Page 7167, lines 4-9: Could this be a function of the uncertainty in BC reported transparently by Bond et al? These differences seem to be very reasonable given the global nature of the Bond et al work cited, demonstrating in fact the opportunity for convergence in the BC inventories as has been achieved over time for most well-studied pollutant emissions. Moreover, despite a difference in Table 1 and Table 2 emissions of

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BC from ships, that span an order of magnitude, there is rather strong signals reported in Table 4 for the region studied that point to the importance of other BC sources even with the uncertainties in ship emissions rates or deposition (as discussed in line 15 on this page).

Reply: Yes, indeed, it is related to the uncertainty of the land based emissions of black carbon. Phrase added: "... which seems to be related to the uncertainty of land based emissions of black carbon, typically a factor of 2 (Bond et al, 2004). "

9) Page 7168, lines 26-28: Do the authors include NMVOC from cargoes or only engine emissions? This could contribute to the discussion, and may help explain the difference.

Reply: In the inventory described by Eyring et al., NMVOC from fuel and loading are included (see Chapter 2 and Appendix A, Table A1a), the emissions from loading account for 50% of total NMVOC emitted. In contrast, other inventories do not consider NMVOC from loading. Without the emissions from loading, the NMVOC emissions in Eyring et al. are nearly same as in EMEP, but the NOx in Eyring is lower. The emissions factor is the main uncertainty for NMVOC.

10) Page 7169, lines 11-15: Again, what would the effect be if cargoes emissions of NMVOCs were included?

Relply: See 9).

11) Page 7170, lines 7-13: I don't understand what the interpolation does for the quality of the analysis, except to imply resolution for illustrative purposes only. This "sharpening" of the resolution is an exercise that does not contribute any definitive information, I think. I would reconsider including it, unless its contribution to the wellcrafted logic of the paper is made clearer.

Reply: We are trying to show that the shipping signal is clearly detectable in the Eastern Mediterranean from the NO2 columns, but we cannot have a quantitative comparison

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with the model because the model resolution of 1x1 degree is too coarse. We believe this is an important statement because we are not able to state the impact of ships on the Eastern Mediterranean from other observations. The OMI data could be possibly used to constrain the ship emissions in Easter Mediterranean in a future study, applying a model with a finer resolution and using EDGARv4 or similar inventories (e.g. Wang et al, 2008) with a resolution of 0.1x0.1 Degree. See Conclusions: "Over the Eastern Mediterranean we find a spatially narrow signal in the satellite NO2 retrievals which is not reproducible by the1x1 model resolution applied in this study. With EDGARv4, we now have a much finer resolved emission inventory which should be applied in a model with a corresponding 0.1x0.1 Degree resolution in future".

12) Page 7167, lines 4-9: Could this be a function of the higher BC attributed to Bond et al for ships? Check the ratios again and compare with Table 4.

Reply: See 8).

13) Page 7171, lines 1-2: The fact that emission factors for NOx and SO2 agree among inventories is not coincidence. This represents significant convergence among independent studies during the past decade. This convergence is even more remarkable for ship inventories at global and regional scales than for other global source categories, and should be highlighted.

Reply: Phrase changed:

"Emission factors for NOx and SO2 agree much better among the inventories representing significant convergence among independent studies during the past decades. "

14) Page 7171, paragraph beginning at line 21: Should start with a sentence that says: "We can also conclude that variation in regional patterns of ship activity, both spatially and temporally, may drive the variation in agreement among satellite, model, and observed concentrations attributed to ships. Then use one of these statements

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(the first is correct, I think): “This is a PRIMARY cause of remaining disagreement among the inventories, except where emissions factor differences are very large (e.g., BC and NMVOCs).” . . . or: “This is a SECONDARY cause of remaining disagreement among the inventories, behind the discrepancies among emissions rates (especially BC and NMVOCs).

Reply: We agree that spatial and temporal variation of ship activity is an important cause of disagreement, but not the primary one. The global inventories analyzed here have the same spatial distribution, and none of the inventories include temporal variation, i.e. they assume an annually constant temporal activity. The spatial variation between the global and the regional inventories as well as the missing seasonality cause important uncertainties, but by far not the main disagreement. The disagreement due to the emission factors, the assumed fuel consumption (based on different methodology) and the total shipping activity (domestic shipping) cause larger differences.. Sentence changed: “Combined uncertainties in the spatial and temporal variation of ship activity, land based emissions, modeled transport and sinks of air pollutants (e.g. wet deposition of BC) determine the variation in agreement among the observations and the model. To illustrate this we present our results in four categories ...” 15) Page 7173, lines 7-9: Please explain the sentence stating “Different seasonal patterns are given by global ship activity data sets ICOADS and AMVER and need more investigation (Dalsøren et al., 2008).” Did your work not consider seasonal variation when reviewing the selected inventories, only cited Dalsoren as evaluating this? Why doesn’t this paper report what the current ACPD study finds? It is important that this paper directly evaluate seasonal differences as an explanatory item in this study, not simply cite another recommending investigation.

Reply: To answer your question about the seasonal representation of shipping activity in the inventories applied: Chapter 2, Ship emissions, page 7162, lines 7-10: “In all investigated inventories a temporally constant emission flux was assumed, i.e. the emissions are constant throughout the year.” Chapter 6, Conclusions and Discussion,

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page 7171, lines 10-12: “Seasonal variability of ship traffic has not been included in any of the inventories; all emissions are assumed temporally constant which presents an additional uncertainty.” In Dalsoren et al. Figure 6 we find a maximum of shipping activity in winter according to AMVER data and a maximum in summer according to COADS data, with maximum variation between the months of 10%. While this is not unimportant, considering the differences between the inventories ranging from 50% to an order of magnitude or more, we do not expect to obtain more accuracy from applying these contradicting seasonal variations to different inventories and find such exercise beyond the scope of this study. We find it important however to mention the results of Dalsoren et al. as an additional source of uncertainty.

Technical corrections:

1) Page 7157, line 8: Editorial comment: Authors could consider updating this with Lack et al, JGR, 2009.

Reply: Reference updated D. A. Lack, J. J. Corbett, T. Onasch, B. Lerner, P. Massoli, P. K. Quinn, T. S. Bates, D. S. Covert, D. Coffman, B. Sierau, S. Herndon, J. Allan, T. Baynard, E. Lovejoy, A. R. Ravishankara, E. Williams, Particulate emissions from commercial shipping: Chemical, physical, and optical properties, JGR, 114, D00F04, doi:10.1029/2008JD011300, 2009.

2) Page 7171, line 14: What is “relative” about this certainty? The paper makes this conclusion certain, in absolute metrics. It is not relative at all that ships impact the region, given that you have used several inventories and all have shown this to be true.

Reply: Sentence changed: “...Our findings show that ship emissions have a significant impact on air pollution over the Mediterranean region.”

3) Page 7159, line 18: Please check reference to Lloyd’s 1999. I believe Erying used a much more current version of Lloyds data - perhaps 2007. This implies many levels of inaccuracies that are not representative of the Eyring inventory (or the Wang et al

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inventories).

Reply: Eyring et al. have used Lloyd's (2002), reference corrected "Lloyd's Maritime Information System (LMIS) (2002), The Lloyd's Maritime Database [CD-ROM], Lloyd's Register–Fairplay Ltd., London."

4) Page 7173, lines 3-5: The reference to biomass burning seems disconnected from the paper, not being discussed at all prior to this as an input tested for sensitivity, but only included among natural emissions "held constant".

Reply: Sentence changed: "The overestimation during summer and the underestimation during winter could also be caused by an incorrect temporal variability of BC from biomass burning, which is an important source in the Mediterranean region (e.g. Palumbo et al., 2006) with a temporally irregular occurrence. "

Reference added: Palumbo I., J. Kucera, P. Barbosa, V.W. O'Brien, R. Valentini, Using SEVIRI geostationary imagery for active fires analysis and burned biomass estimation: A case study in a Mediterranean ecosystem, Forest Ecology and Management, 234, S222, ISSN 0378-1127, DOI: 10.1016/j.foreco.2006.08.249.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 7155, 2009.