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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.

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Answer to reviewer 2, Dr. Mark Lawrence

Thank you for your helpful review. We agree with most of your comments and have revised our manuscript accordingly. Please find below a detailed overview of our revisions.

1) The differences between the emissions datasets should be stated more carefully throughout, e.g., qualifying that the two orders of magnitude difference noted in the abstract only applies on a very limited basis, while the important emissions on even a small regional basis are generally quite consistent.

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.

The sentence has been therefore changed to: "... in a difference ranging from a factor of 1.5 to even an order of magnitude". See our comments to Reviewer 1.

2) It would be very valuable to the study if it were possible to carry out a sensitivity simulation with an assumed temporal variability in the emissions, quantifying at least the likely range of impact of this, rather than just indicating this is a need for future studies.

Reply: In Dalsoren et al. (2008) 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 significantly different results from applying these -contradicting - seasonal variations to different inventories. Sentence added: "We decided not to perform further sensitivity studies on this item die to the expected low signal and in order not to expand the length of the manuscript."

3) This work should be placed better in the context of past literature. Dr. A. Richter pointed this out for satellite studies in his comment; in addition, the results here should be put in perspective of the results from seminal papers on this topic, particularly Ca-

paldo et al. (Nature, 1999) and Lawrence and Crutzen (Nature, 1999), and the few major developments since then; beyond these, in the abstract it is stated that "very little has been done to evaluate their consistency with atmospheric measurements at open sea", which neglects the fact that a comparison to aircraft observations was already included in Lawrence and Crutzen (1999), and a much more detailed comparison (leading to a debate about the overall significance of ship emissions for NOx) was focused on in Kasibhatla et al. (GRL, 2000) and Davis et al. (GRL, 2001), and similar comparisons are available for other species.

Reply: We agree that a lot of work has been done to evaluate ship emission estimates and we regret the serious omission in the manuscript. 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), Kasibhatla et al., (2000), Davis et al., (2001) and Lawrence and Crutzen, (1999) have included ship emissions into chemistry transport models to evaluate them with aircraft measurements over Pacific and North Atlantic. Wand et al. (2008), Corbett et al. (2008) and Lack et al. (2009) 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). Franke et al. (2008) combine 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."

4) The satellite comparison should also be placed in the context of the uncertainty in the satellite retrievals; I am not very familiar with OMI, but for GOME and SCIAMACHY often a detection limit of about 10ËĘ15 molec/cmËĘ2 is assumed, and the observed values in Figure 11 are not much above this; a brief discussion of the OMI uncertainty should be included (perhaps in an appendix), as well as a clear indication in the text of what "weak but significant signal" means, and that this error will be in addition to the

(already relatively large) standard deviation bars included in Figure 11.

Reply: In section 4.2 we discuss our best estimate of the error on a single OMI tropospheric vertical column which amounts to 1.0 10ˆ15 molecules cmˆ-2 from spectral fitting. However, this uncertainty is strongly reduced by temporal and spatial averaging. For a 3-month average at 1ËŽ×1ËŽ (and over the complete Mediterranean region as in Figure 12), each grid contains on the order of 500-1000 cloud-free OMI observations, so that the random error is small (typically less than a few percent). We no longer state that a weak but significant signal can be recognized, but rather that the most prominent feature in Figure 11a is the elevated (by ∼1.0 10ˆ15 molecules cmˆ-2 relative to background levels) NO2 along the main ship route extending from the Street of Gibraltar to Egypt. We have updated the text in section 4.2 and 5.3 accordingly.

Text revised: "Here we focus on summertime observations, when cloud free conditions prevail. Each grid cell contains 500 to 1000 cloud-free OMI pixels and the random error is significantly reduced by the spatial averaging".

"Figure 11a shows the average OMI NO2 tropospheric columns (gridded to 0.125x 0.125) over the Mediterranean North America for June-August 2006. The most prominent feature in Figure 11a is the elevated (by ∼1.0 10ˆ15 molecules cmˆ-2 relative to background levels) NO2 along the main ship route extending from the Street of Gibraltar to Egypt".

As indicated above, the uncertainty on the JJA average at 1x1 is only a few percent, so that the standard deviation will be dominated by geographical variability in NO2 over the Mediterranean domain (Figure 12 shows spatial variation).

5) Finally, the results should also be placed in the context of what is already known about the behavior of TM5 from other studies and evaluations compared to global observations; this is particularly relevant for BC, which is found here to be overestimated even in the no-ship-emissions simulation, but also applies to NOx and O3, helping the reader to calibrate the extent to which the TM5 simulation is known to reasonably

simulate global atmospheric chemistry (depending on how it compares, e.g., in the Stephenson et al. and Dentener et al. intercomparisons, this might end up helping strengthen the overall conclusions).

Reply: The overestimation of black carbon in summer is not very significant, it is less than 10% without ships, and the agreement improves close to the main shipping route (Tunis-Palma leg, Table 4). Much more significant is the underestimation during winter over the sea as well as in Finokalia (30-50%).

Text added: "TM5 has participated in a number of recent global model intercomparisons, such as PhotoComp (Dentener et al., 2006, Stevenson et al., 2005) and AEROCOM (Kinne et al., 2006 and Textor et al., 2006). TM5 generally performed among the better global models regarding ozone and oxidized nitrogen depositions, probably due to its relatively fine horizontal resolution. TM5 also participated in the recent regional EURODELTA exercise and was found to perform reasonably well with regard to ozone surface concentrations in Europe with good correlations in summer and winter (van Loon at al., 2007). A good agreement of simulated surface NOx with EMEP measurements in winter and summer was found by deMeij et al. (2006) while black carbon was found to be underestimated by the model especially during winter by a factor 2.5 on average."

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