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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global distribution of sea salt aerosols: new constraints from in situ and remote sensing observations" *by* L. Jaeglé et al.

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General Comments: The authors present a new formulation for the sea-spray source function that takes into account that water temperature affects the production of sea-spray particles. The formulation is based on the comparison of sea-spray aerosol concentrations obtained from five cruises in different parts of the world covering the major oceans and sea-spray aerosol concentrations computed with the GEOS-Chem model using the Gong (2003) formulation for the sea-spray source function. The question arises whether this is the most appropriate source function since it is extrapolated from the Monahan et al. (1986) source function to smaller particles, using experimental data from O'Dowd et al. (1997) but for sizes smaller than those for which data are available. Also the large size limit of the Gong (2003) source function is larger than

that given by Monahan et al. More recent source functions exist, e.g., from laboratory experiments by Mårtensson et al. (2003) and surf zone experiments by Clarke et al. (2006). These latter source functions are in reasonable agreement and comparison of these with Gong (2003) show clearly that Gong falls off to fast at the small particle end. This is important for the smallest particles considered in the work by Jaeglé et al. of rdry = 0.01-0.5 um (25696, 9). The authors should explain their choice for Gong et al. and evaluate the problem at both the small and large particle end.

Indeed, many different sea salt source functions exist. Our approach was to use the source function that showed the smallest initial bias with respect to in situ and remote sensing observations, and then examine the remaining bias as a function of SST. Using the Clarke et al. (2006) source function in GEOS-Chem leads to very high emissions of sea salt (five times larger than Gong for coarse mode sea salt and twice as large as Gong for accumulation mode sea salt). This is similar to the results of Pierce and Adams (2006). When compared to PMEL observations, we find that the Clarke et al. (2006) source function leads to a large overestimate of PMEL sea salt mass concentration observations: +95% for accumulation mode and +500% for coarse mode (compare to +7% and +64% for Gong). The Martensson et al. (2003) formulation extends up to 1.4 μm, so we cannot use it to examine coarse mode sea salt. Accumulation mode sea salt aerosol mass concentrations measured by PMEL are overestimated by a factor of 2 by the Martensson et al. (2003) source function. The Monahan and Gong formulation are very close, but do differ at the small particle sizes (in terms of mass for rd<0.5 um Monahan is 25% larger than Gong) and large particle sizes (for rd>0.5 um Monahan is 10% smaller than Gong). For completeness, we now include a comparison of the PMEL observations to all four sea salt source functions in the supplementary material.

The comparison of model results and experimental data shows that the model concentrations are much too high. The authors first investigate whether the wind speed *dependence used in the model fluxes would be too high and propose an alternative formulation for the sea-spray source function with both a much smaller wind-speed dependence and different coefficients (MODEL-U2, eq 3). However, although this new formulation brings observation and model closer together this correction is not sufficient. The evaluation of MODEL-U2 is only briefly discussed in section 4.4 and the focus of the rest of the paper is on the sea surface temperature (SST) dependence. I suggest that the conclusion on the less good performance of MODEL-U2 is included in Section 7 or 8.*

We have added a discussion of MODEL-U2 in section 8.

The ratio of experimental and model concentrations appears to be clearly dependent on SST and a third order polynomial fit of this ratio to SST is included in the sea-spray source function formulation (MODEL-SST, eq.4). The use of MODEL-SST brings the model results much closer to the observations. This is not surprising since the same observations were used in the development of the SST dependence and it's evaluation. I suggest that the authors mention this in the manuscript and that the real evaluation of the MODEL-SSA is in the comparison with the UniMiami ground-based network data, the MODIS AOD and the AERONET results.

Indeed, the ground-based observations and the MODIS+AERONET AOD observations provide an independent evaluation of MODEL-SST. We have added a sentence emphasize this at the end of section 4.5

Detailed comments.

Title: hyphenate sea-salt

The Oxford and American Heritage dictionaries do not hyphenate sea salt. The current scientific literature on the topic of sea salt aerosols is split, some authors hyphenate it, while others do not. We have decided to keep spelling it as sea salt, without the hyphen.

25701, 3-13: dry vs wet deposition: It is a bit surprising that the removal of data influenced by precipitation does not influence the comparison. This would imply that precipitation is very well modeled. How is wet deposition included in the model?

We exclude observations affected by rain to see whether model errors in wet deposition are causing the discrepancy between model and observations. Even when we exclude these points, the bias remains. Similarly when we only consider data points influenced by rain the bias remains. This only means that the bias exists both when it rains and when it doesn't rain. This points us to emphasizing potential biases in sea salt emissions instead of sinks, even though considerable uncertainty remains in terms of different parameterizations of dry and wet deposition of sea salt aerosols. The text has been modified as following: "We further examine the model bias for coarse mode SS by excluding observations affected by rain. This greatly reduces the number of available points (from 383 to 134) but does not change the level of disagreement. The bias also persists if we only consider observations affected by rain during the sampling period. We use this to argue that model errors in the representation of wet or dry deposition cannot explain the poor model performance. Dry deposition dominates the loss of coarse mode SS and considerable uncertainty remains on the calculation of vd. Lewis and Schwartz (2004, p. 277-283) assess different model formulations for vd, finding that they generally agree to within a factor of 2. This is a smaller range of uncertainty than that associated with SS emissions (factor of 4). In the rest of this paper we thus focus on potential biases associated with the SS source function, its wind speed dependence and its dependence on other variables. "

25701, 14-eq (3): the observations are local, the model considers an, in comparison, very large grid of 2x2.5 degrees. Are variations across the grid accounted for and how?

No, we do not account for any subgridscale variations in sea salt concentrations

because of the global nature of our model. We thus inherently assume that the local sea salt mass concentration observations reflect a larger surrounding region. We also note in our revised manuscript that: ”The sampling period of the impactor ranged from 2 to 24 hours, with a mean of 11 hours. With an average ship speed of 13 knots (24 km h-1) this corresponds to measuring SS concentrations over 50-580 km, with a mean of 265 km. This scale is comparable to our model grid-size (200-300 km).”

Figure 4 shows ratio and r, I suggest to also provide the regression lines (intcpt, slope) to help understanding the further analysis, in particular the derivation of eq. 2. Cobs/Cmodel would be simply given by the slope (is that what's meant with ratio in the Figure legend?) since Fig 4 suggests a straight line, and Eobs would be simply a multiplicative factor times Emodel. However, if that is true, I don't understand how the wind speed dependence would change. Apparently the Cobs/Cmodel ratio has a non-linear dependence on wind speed, but that's not clear from the text nor from the Figures. I assume that eq (2) is evaluated for each individual data point to get to Figure 5. Could you please clarify the issues raised here in the text?

In order to derive Eq (2) we assume mass balance for coarse mode sea salt aerosols in the surface layer. This yields a linear relationship between E (sea salt emissions, μg m-2 d-1) and C (surface sea salt mass concentrations, μg m-3): E = $\alpha \times$ C, with the linear coefficient α depending on the residence time of surface SS, τSS, and the depth of the surface layer (zLS): α = zSL/τSS. In practice, we calculate α with the GEOS-Chem model as α = Emodel/Cmodel. We can thus use the model to infer Eobs = (Emodel/Cmodel) $\times$ Cobs, which is Eq (2) in the text. This equation is evaluated at each point to infer Eobs and the results are plotted in Figure 5 (left panel). Our assumptions in deriving Eq (2) have been clarified in the revised manuscript.

25704, 15: Sellegri et al. (2006) also addressed water temperature dependence

We have added a citation to this paper.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C13343/2011/acpd-10-C13343-2011-supplement.pdf

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