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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Primary marine aerosol emissions: size resolved eddy covariance measurements with estimates of the seasalt and organic carbon fractions" *by* E. D. Nilsson et al.

Anonymous Referee #1

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Nilsson et al. present size resolved measurements of coastal aerosol fluxes with a set-up that can differentiate between the total flux and the flux of non-volatile components of the particles. The study concludes that the flux of the non-volatile component shows size and wind speed dependences that match recent sea salt emission parameterisations. Interestingly, however, the total particle flux is clearly higher and not dependent on wind speed below 10 m/s. The authors claim this fraction of particles to primary organic components from an emulsion on ocean water and emitted as an internal mixture with sea salt.

All in all, the paper addresses several important scientific questions and provides new

data that add to our understanding of both sea salt and organic marine sources. While my comments and criticisms regarding the methodology and results of the study are fairly minor, I do not fully understand how the authors have reached some of their conclusions. Furthermore, the language of the manuscript must be thoroughly revised. The numerous errors in grammar, spelling, use of vocabulary etc. on each page along with the very wordy style of writing make the paper laborous to read. At the very least the manuscript must be carefully checked and corrected by a native English speaker but I also strongly recommend that the authors take some time to condense it by reducing the speculative and repetitive parts and by concentrating only on details directly relevant to the study at hand. After the specific comments, listed in detail below, have been addressed and the language improved, I recommend the publication of the manuscript in ACP.

Specific comments:

p. 13348, l. 20-24: Does "below ˜100 nm diameter" refer to ambient (wet) diameter? Please specify. In remote marine conditions the minimum *dry* activation diameter can easily be ˜50 nm even for relatively low updraughts ˜15 cm/s. In this size range also secondary sulphate particles entrained from FT are important and thus can affect CCN (e.g. Spracklen et al., ACP, 2007). I therefore disagree with the authors' statement that primary marine aerosol determines the (CCN) number concentration; rather, it is the combined effect of primary and entrained secondary particles. It is true that secondary sources have also another effect on CCN: they grow pre-existing particles (both sea spray and sulphate) into larger sizes as concluded by Pierce and Adams (2006). However, their model uses a fixed activation diameter of 80 nm as well as a nucleation mechanism that is likely to underestimate the nucleation rate in FT, both of which probably cause underestimation of the effect of secondary entrained particles from FT on CCN.

subsection 1.2: The description of the sea spray parameterisations should be reduced significantly. This goes for all three parameterisations but especially the text on MO86

ACPD

7, S6114–S6120, 2007

and GO03 is very long and contains information that is not directly relevant to this study. For MO86 and GO03 it is sufficient to state the correct measured range and RH, *briefly* mention that the parameterisation is commonly used outside this range, the need for Gong extrapolation, and the fact that GO03 has a freely adjustable tuning parameter that makes it unreliable. All this can be said in 10-15 lines. I do understand that the authors want to promote their own (and indeed very good) parameterisation but I find the style of the description of the other parameterisations slightly patronising.

p. 13357, l. 4-26: Much of the text can be reduced without losing the main points that current measurement height is above internal BL and that the chosen sector is close to open sea conditions.

p. 13358, l. 16: Do the authors mean eqs 4 and 5? If not, please explain how eq. 3 used in correction.

p. 13359, l. 15-16: What does "increase to $<$5%" refer to? The previously mentioned fractions are *higher* than 5%. It is also unclear what "0.1-0.3 and 0.1-1 um" refer to? Do the authors wish to say that below 0.3 um for the sea salt and below 1 um for the total aerosol the penetrating factor is $>$90%?

p. 13360: - l. 5-8: What effect do the very large errors at large particle sizes have on the results and conclusions? - l. 12-15: Not relevant to this study, please delete. - l. 17-20: Even if neglecting sedimentation doesn't affect *net* aerosol flux much, this study focuses on size-resolved fluxes - how big is the effect on these? - l. 23-28: Oddly placed and unnecessary paragraph. The sentence "The errors presented in Figs. ..." can be moved to subsection 2.5.1. (assuming "Eq (5)" should be eq. 6 for discrete counting). Please delete the rest.

p. 13361 - l. 3-13: Not necessary to understand the results; please delete. - l. 20-24: CPC measurements can also indicate that ultrafine particles had sources other than the sea surface, such as entrainment from FT.

ACPD
7, S6114–S6120, 2007

EGU

p. 13362 (and later in text) It is difficult to follow when heated and unheated OPC measurements are discussed. Please make this explicit in the text. Furthermore, using the term "total aerosol number flux" here is confusing."Total aerosol" does refer to the unheated aerosol (according to authors' earlier definition) but not in the same size range as the OPC measurements. Thus the factor 5 difference between heated OPC and CPC measurements is caused by two factors: different size range and heating. What does this comparison then tell us? On second to last line these two fluxes are referred to "$>$11 and 100 nm diameter" - again indicating that they would be comparable in all other aspects apart from lower size limit.

p. 13362, l. 12-18: The text suggests that MA03 agrees better with CPC measurements than CL06. This is true for some periods and untrue for others. There certainly are periods when CL06 underestimates CPC measurements - but for most of these periods so does MA03. There are also periods when CL06 is spot on and when MA03 overestimates the flux. If the authors wish to make a statement about the relative performance of the two parameterisations, they should do so over a much longer time period (here 24 h!) and use quantitive measures.

p. 13363, l. 6-9: Why do CPC measurements anticorrelate with wind speed?

subsection 3.3.1: Fig. 4 shows fluxes for four 30 min. periods with different average wind speeds. How much variation was there in wind speed *within* each of these periods, i.e. is the average wind speed the right variable to look at (the flux was found to follow a power function, so it doesn't scale linearly with wind speed)? Can these periods be considered representative (and why?) or should the plots be given for all/many more periods with the same wind speed (this also goes for Fig. 7)? Please give the measured sea water temperature; assumingly it was used for MA03 in Fig. 4. I do agree that CL06 and GO03 seem to systematically underestimate the flux for the smallest size. However, apart from 4c, all the parameterisations capture the flux ˜0.8 um and above badly, not only CL06. For size range 0.3-0.8 um there is quite a bit of fluctuation between subsequent size ranges and it is not immediately clear which of the

ACPD

7, S6114–S6120, 2007

parameterisations performs "the best". The measurements do not show a clear slope so such a slope should not be used for comparison.

p. 13364, l. 16-22: Is the median calculated for each size bin separately or is it the actual median distribution?

p. 13365: Figure 5 itself is interesting and relevant but many of the main conclusions based on it have already been presented in earlier sections so the text can be significantly reduced. - On lines 1-14 there is repetition of results presented earlier; MA03 temperature dependence has been studied in Martensson et al. (2003); it is sufficient to say that because of the temperature dependence of flux CL06 is not valid outside the tropics i.e. please delete 11-14. - MA03 at 25C and CL06 do not agree at all below 200 nm or above 1 um. How can CL06 be said to validate MA03 for tropical waters? - Lines 15-29: The main result (GO03 doesn't match observations [=current study & CLO6 - not MA03!] regardless of the value of the tuning parameter) can be presented in 5 lines. Please reduce.

p. 13366: Fig. 6 and its caption do not match at all. Please delete lines 8-15. The lowest size channel at 100 nm does not agree well with any of the percentiles.

p. 13367: - l. 4: No need to refer to Fig. 4 when the same information is discussed in the figure the text is talking about. - l. 5-6: Fig 8. does not show volume fluxes as indicated by the text (also referred to on lines 13-14), although such a plot would be very interesting to see.

section 3.5: Please switch the order of second and third paragraphs. In (currently) third paragraph: is there any experimental or theoretical support for the hypothesised rupture of organic film/emulsion at high wind speeds? If not, it is ok to speculate here but not in the abstract.

section 3.6 and Fig. 9: Can be deleted altogether as Figs. 4-7 and the related text give exactly the same information. Discussion of biological activity can be moved to section

ACPD

7, S6114–S6120, 2007

3.4.

section 3.7 Intuitively the authors' conclusion the they observe internally mixed particles seems correct - after all it is difficult to imagine a process that creates pure organic particles and has the same wind speed dependence than the sea salt flux . There are couple of points in their reasoning, however, that need clarification. - Fig 10: There is a clear rise in concentration in the smallest size range at 100 nm (seen both in number and volume). This is not seen in the shifted flux at 100 nm. Is there a reason to doubt the measurement accuracy at this size chanel (it doesn't match MA03 number flux either)? If not, could this rise be an indication that the particles in fact are externally mixed, especially if the downward shifted total flux matches the sea salt flux? - p. 13370, l. 16-17: Is factor 3.7 based on average or median fluxes? - p. 13371, l. 4-8: Wouldn't a better example to fig. 8 be considering particles sized *initially* 100 nm, which after losing their organic coating would decrease below 100 nm and thus the measurement range of OPC? - p. 13371, l. 15-17: Why does this matter? Isn't the only the corrected (i.e. "correct") flux that needs to match if particles are external? - p. 13371, l. 18-24: Could the hypothesis of Leck and Bigg (2005) for the Arctic be relevant here? They claim that at emission particles are internally mixed organics and sea salt but that these two components separate in the atmosphere and organic component goes through subsequent break-up due to UV radiation and acidification. This would be (at least approximately) consistent with the same wind speed dependence of total and sea salt fluxes. - Wouldn't an organic layer on sea water/in droplets affect the bubble bursting (e.g. through surface tension effects)? I find it intriguing that the sea salt part of the internally mixed particles (if organic assumed to be of primary origin) follows exactly the flux measurements made with synthetic sea water.

section 3.8: -This section is extremely long for a purely speculative hypothesis, although the hypothesis itself is interesting and can in my opinion be presented in a much condensed form (in max. 2 pages instead of current 6). It is sufficient to briefly introduce the Ellison model and the uncertainties related to it, and then to describe

ACPD

7, S6114–S6120, 2007

how the combined MA03 and Ellison model performs against observations. It is impossible to draw any conclusions based on this exercise without more concrete evidence of the mechanism. - subplots b and c in Fig. 11 are in odd order compared to all other figures. - On p. 13372, l. 14 it is said that sea salt volume is 2% of total volume. This contradicts with Fig. 11 and thus the match between the model and measurements cannot be considered very good.

Summary and conclusions: - point 3: This conclusion must be rewritten. MA03 is not quite as superior as the authors like to think (see comments above). - point 4: This conclusion needs to be reduced and rewritten to illustrate that Ellison model is not currently validated. I also disagree with the last sentence as analysis from many other sites is needed to validate this statement (same with last sentence of abstract).

Technical comments:

Please change "full curve"/"full line" to "solid curve"/"solid line".

Please change "Julian day" to "Day of year" or "DOY". Julian day most commonly refers to day lapsed from beginning of year 4713 BC, i.e. current Julian day would be around 2.5 million.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 13345, 2007.