

Interactive comment on “Model study on the dependence of primary marine aerosol emission on the sea surface temperature” by S. Barthel et al.

Anonymous Referee #3

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1 Outline of the manuscript

Barthel et al. present an evaluation of primary marine aerosol (PMA) emissions from the atmospheric transport model COSMO-MUSCAT using measurements of sodium concentrations obtained during two intensive EMEP campaigns as well as Berner-impactor measurements obtained at Sao Vicente (Cape Verde). As per the title of the manuscript the major emphasis of the paper is evaluation of the PMA source function that they employ in their model, that presented by Long et al. (2011), in the context of sea surface temperature (SST). To do so the authors test the application of a num-

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ber of different correction factors designed to account for the effect of SST on the PMA source function. These correction factors are derived either from previous PMA source parameterisations that incorporate a SST dependency (e.g. Jaeglé et al. (2011) and Sofiev et al. (2011)) or they parameterise laboratory generated sea spray fluxes generated at different SST's (Zábori et al., 2012).

Unfortunately, I find little of any scientific value in the manuscripts findings and question the novelty of the work. I also have a series of serious reservations regarding the approach used by the authors to the extent that their conclusions may be unfounded. Thus I must recommend that this manuscript is rejected in its current form. For the sake of transparency I will also add that I did not have time to review the manuscript at the “quick review” stage as the manuscript was accepted prior to the deadline date set initially. If I had been able to submit a review at this stage I have no doubt that my conclusions would have been the same.

2 Major points

As written, the manuscript has many major methodological problems:

- The authors have chosen to use the PMA source function of Long et al. (2011) as their baseline parameterisation. Not only do the authors justify this decision based upon comparison to only a single station, seemingly ignoring comparison to all the other stations described in the manuscript, they also fail to include any depiction of how good the agreement between model results and observations was. They must turn what is currently a qualitative justification of their choice based upon a single station into a quantitative assessment based upon all the data at their disposal.
- The measurement data used in this work are primarily the concentrations of

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sodium (Na⁺) in air obtained during two EMEP intensive measurement periods in June 2006 and January 2007. Here low volume samplers equipped with quartz filters were typically used to collect PM₁₀ and PM_{2.5} (or PM₁). The PM samples are then used for weighing and subsequent chemical analyses. Such samplers employ an aerosol cut-off at ambient relative humidity (RH) which given the high hygroscopicity of sea salt aerosol means that the actual size cut-off of the sampler will be highly dependant on RH. In Figure 1 an example of the hygroscopic growth factor of NaCl is given. At elevated RH (not uncommon at coastal stations in the marine boundary layer (MBL)), the actual size cut of the dry particle diameter (D_p) decreases substantially for very hygroscopic particles like sea salt. Under these conditions, the largest particles able to enter the PM₁₀ sampler will be of significantly smaller D_p (e.g. a PM₁₀ size cut is effectively reduced to a PM₅ size cut, etc.). If such data are then compared to the model results a significant mass underestimation will occur. This will apply to all size fractions of the measurement data (i.e. PM₁ and PM_{2.5} also) and undermines the conclusions of the paper given that they are based upon mass differences between the observed and modelled data. I suggest that the authors apply some cut-off to their data, perhaps remove all days where the ambient RH at the EMEP station exceeds RH=80%. That or they could attempt a similar approach to that adopted by Gryth et al. (2014). This will significantly decrease the uncertainty here.

- Given the authors apparent lack of attention to the quality of the measurement data, a subsequent question which arises is whether there are any quality issues with the data? No mention of quality assurance is made in the observation section of the methods and discussion of whether any data has been removed for quality purposes is absent. Given that only a few months of data from a few stations are used this is a necessity.
- Another issue concerns the authors choice of observation stations. As well as choosing some coastal stations to compare to they choose a number of stations

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which are at best questionably coastal and some which are distant from any ocean. In these instances are the authors testing a source parameterisation in their model or its transport and deposition dynamics? This obscure approach dilutes the message of the paper which according to the title is the effect of SST on the source of sea spray. These stations should be removed.

- Following on from the previous point I would also like to see some basic trajectory analysis deployed. Just how many days at each station were actually likely to contain significant amounts of sea spray? How does the model compare with measurements on these days compared to those less influenced by sea spray?
- The next issue is whether sea ice is included in their model. I could not find any description of this in the methods section so assume that it is not. This is a flaw given the focus on SST and sea spray emission, especially when the correction factor of Zábory et al. (2012) is applied. This dataset implies that the source of sea spray will increase significantly at low SSTs, thus the difference between an SST of 0 °C and a region covered by sea ice will be stark. This problem is illustrated by the Virolahti data. The authors note lower sea spray here in the observational data when compared to their model. The authors reason that this effect is entirely due to the low salinity of the Baltic Sea. However, it is clear that the sea spray source region for this station is likely to be highly influenced by sea ice in the winter. Thus, whilst the authors reasoning on salinity effects may well be important, concrete conclusions cannot be drawn.

The manuscript also has significant presentation flaws in its current state:

- The manuscript presents some comparison of organic sea spray emissions. However, given the almost complete lack of discussion of this data (limited to a couple of lines in the discussion) this only seems to dilute the take home message of the manuscript which is the effects of SST on PMA. This should either

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be expanded upon significantly (thereby changing completely the focus of the manuscript) or be completely removed.

- Many of the figures are illegible in their current form when printed. I cannot comment adequately if I cannot read the axis labels. Further to this many of the figures are likely redundant. A single set of nicely presented examples followed by presentation of regression stats in a table (regressions between the observed data and modelled data) would suffice and make it much simpler for the reader to see how well the source functions/correction factors fit the observed data.

3 Minor points

Given that the minor points are too numerous to outline completely here I shall provide only a few examples to aid the authors:

- Please refrain from unreferenced speculation in the introduction and limit it to the discussion. For example on P382 Line 22 “At higher wind speeds the concentration of organics in PMA is lower due to decreased near-surface concentration through stronger oceanic mixing” should either have a reference or be removed.
- P384 Line 2 The statement “The resulting higher residence time in cold waters leads to an increase in the coalescence of bubbles, thus decreasing the number of smaller bubbles and increasing the number of bigger bubbles (Pounder, 1986)” is controversial. To date no systematic study of the effects of temperature on sea-water bubble concentrations has been reported, and there have been conflicting reports from the few investigations on the topic (see P.249 of Lewis and Schwartz (2004)). I suggest either removing this statement completely or a more thorough discussion of the range of conflicting results observed.

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- Throughout the manuscript when stating particle sizes please remember to state the RH. As a couple of examples P384 Line 24 when you state “for particles with diameter between $0.012\mu\text{m}$ and $1.8\mu\text{m}$ ” do you mean dry diameter or at some RH? Again on P392 Line 3, please be precise and mention the RH.
- A number of sentences need rephrasing to add clarity. For example the sentence on P384 Line 24 makes little sense to me, do you mean it to read “Mårtensson et al. (2003) identified increasing particle production with decreasing SST up to a size of $0.1\mu\text{m}$ dry diameter. The rate of this production increase with decreasing SST was dependent upon particle sizes with the biggest increases at the smallest particle sizes measured.”

4 References

Grythe, H., Ström, J., Krejci, R., Quinn, P., and Stohl, A.: A review of sea spray aerosol source functions using a large global set of sea salt aerosol concentration measurements, *Atmos. Chem. Phys. Discuss.*, 13, 20729-20781, doi:10.5194/acpd-13-20729-2013, 2013.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 14, 377, 2014.

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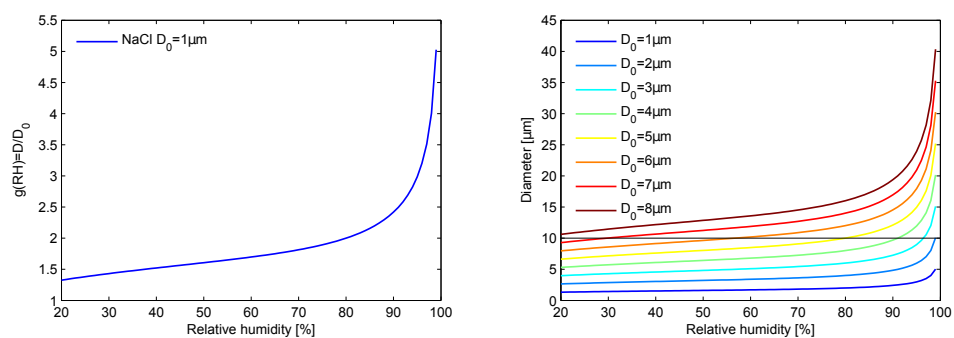


Fig. 1. Left: The hygroscopic growth factor for pure NaCl vs. RH (D_p of $1 \mu m$). Right: The resulting diameter change for different dry diameters. The black line indicates as an example the PM10 size cut.