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Atmospheric Chemistry and Physics Discussions

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

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We thank the reviewer very much for the critical reading of the manuscript and naming some critical remarks, which led to an improved article. We tried to include the remarks by the reviewer into the revised version of the manuscript. Below the review is summarized and our responses to the remarks of the reviewer are presented in indented and italic.

The motivation for selection of the observation stations is not clear. Fig. 1 below shows geographical locations of measurement stations used for the model evaluation. Why

were stations so far inland selected for the evaluation of PMA emissions schemes? How well was the transport and deposition captured by the model? How was the data selected? What are the accuracies of the measurements? Was there any back trajectory analysis done? Overall, there is so little information given that it is impossible to even evaluate the relevance of the data for the model evaluation.

- We spreaded this point into a few single points within the next lines:

The motivation for selection of the observation stations is not clear. Fig. 1 below shows geographical locations of measurement stations used for the model evaluation. Why were stations so far inland selected for the evaluation of PMA emissions schemes?

- (See also response to reviewer 3) Except Melpitz all used stations are less than 40km inland from the sea (Virolahti II <10km, Birkenes <20km, Auchencorth Moss <40km and Cabauw <40km). There are a several reasons why we used these stations: The model simulations were carried out with a horizontal resolution of 28km. Since all named stations are less than 40km away from the coastlines they are within the first or even the second grid cell from the sea. In these cases the influence of the deposition within the model is small. When the distance to the sea is less than 20km such a station may even be within the same grid cell as the coastline. Therefore we assume the error made by using these stations is small compared to other uncertainties. In all model simulations the description of transport and deposition processes is kept unchanged. The aerosols are only tracers and have no influence on the dynamics of the model. Changes in the source strength of the aerosols do not influence the transport properties. Due to that, a change in the emission rates will lead to changed concentrations at all measurement sites, also far inland (Melpitz). This allows conclusions on the sources as well. However, given the uncertainties in the transport- and deposition description within the model conclusions on PMA sources should only be derived from coastal stations. Results from the inland station Melpitz are provided as

additional information, in particular for the PM1 size range that is not available at most coastal stations. The coastal station Mace Head would be ideally representing marine air masses. However only the PM2.5 and PM10 concentrations for January 2007 were available and not for June 2006 (Darius Ceburnis, pers. comm.). Therefore the station could not be used in this work to show the difference between summer and winter situation. Further stations that are located directly at the coast were not available for that time and domain.

How well was the transport and deposition captured by the model?

- The model system has been used in several model studies on aerosols [Heinold et al. 2011a, Heinold et al. 2011b, Heinold et al. 2012, Hinneburg et al. 2009, Renner and Wolke 2010, Wolke et al. 2012) (since 2009) as well as a comparison of the modelled dust deposition to measurements at Cape Verde (Niedermeier et al. 2014). Due to that a further discussion on the model performance regarding transport and deposition was considered not necessary. Nevertheless we extended the number of references for the model performance. In addition, and also due to a comment by reviewer 2 we added a short comment on the deposition: " The comparison of the PM1 and PM2.5 concentrations to the measurements at different stations indicated that the deposition rates may be overestimated by the model. The dry deposition parameterization used in the COSMO-MUSCAT model may overpredict the deposition rates within the PM1 and PM2.5 size ranges (Kouznetsov and Sofiev 2012). Using a dry deposition scheme, which results in lower deposition rates would lead to the aerosols being transported farther inland within the model. That would enhance the role of the effect of the SST on PMA concentrations inland. However the model setup using the current dry deposition scheme has been used in several model simulations obtaining good results (Heinold et al. 2011a, Wolke et al. 2012, Niedermeier et al. 2014). The role of dry deposition parameterization for the modelling of PMA concentrations will be investigated in a future study."

How was the data selected?

- The choice of the measurement period and observational data was due to their availability. Due to the intensive measurement campaigns at the EMEP-sites more stations with measurements of sodium concentration in PM1, PM2.5 and PM10 were available compared to other time periods. A further reason for selecting the measurement sites was their location. Except Melpitz all stations are relatively near to the coasts. Since there were no other real coastal stations available they were the best choice for this work.

What are the accuracies of the measurements? - (See also response to reviewer 3) The measurements in Europe had already been discussed in Manders et al. 2010 (same stations, earlier data), Tsyro et al. (2011) and several EMEP-reports like Yttri et al. (2008) and been used for sea salt model study by Tsyro et al. (2011), who also focussed on the uncertainties. Therefore a further discussion on measurement uncertainties was considered unnecessary. For the station at Sao Vincente a discussion on the measurements and their quality is included in Müller et al. (2010). Here all data were included from that station as no critical values were found.

Was there any back trajectory analysis done?

- (See also response to reviewer 3) Within the revised version we included backward trajectories. These trajectories are not directly shown in the revised manuscript. They are included indirectly within the time series figures 6-8 by different point types. Within the analysis the trajectories have also been included by showing additional plots for data where the air mass came nearly directly from the sea taking less than 6 h. The results are discussed with respect to the trajectories: "Like PM2.5 the sodium concentration in the PM10-PM2.5 size range shows no clear difference between the six-hour and the all-data results. [...] Especially for winter Zb13 leads to overpredictions. The strongest overprediction exists when the air masses reaches Virolahti II from the northwest. Those air masses were influenced by emissions from the Baltic Sea that was at near-freezing temperatures in January 2007."

It is not clear why Long et al. (2011) parameterization was selected for modelling marine organic matter (OM). Authors quote a paper by Gantt et al. (2012) that shows that Long et al. (2011) parameterization did not capture seasonality of marine organic aerosol at Mace Head and Amsterdam Island. Also, when using Long et al. (2011) parameterization, model-predicted surface concentrations showed zero or negative correlation with monthly- and weekly-averaged observational data (Gantt et al., 2012). Recent paper by Rinaldi et al. (2013) gives further insight into organic matter enrichment in submicron primary marine aerosol. It should be noticed that Long et al. (2011) parameterization yields very high enrichment of organics in sub-micron fraction. For example, for [Chl-a] = 0.3 and 1.0 Eq. (12) in the current paper yields RV,1 between 1400 to 0.3 and 2200 to 0.33, respectively for 0.01< Dp <1 μm. Under such high organic enrichment (see Fig. 11 in the current paper) it is hard to argue "the influence of OM on water uptake can be neglected." The assumptions used in the current paper on hygroscopic growth of sub-micron PMA can lead to considerable uncertainties.

- The Long et al. (2011) - OM-parameterisation was used because it was provided together we the sea salt source function that was used here. The results from this parameterization agree quite well with the OM measurements at Cape Verde. The comparison was done for two months in summer and in winter. Since the focus of this paper is not OM, we decided to show only the month with the more measurement points. We point out the discrepancy to Gantt et al. (2012). While Gantt et al. (2012) emitted organic material additional to sea salt we treated both to be internally mixed and emitted together as PMA. The total volume emission rate is kept unchanged due to the presence of OM in our approach. While Gantt et al. (2012) found the Long et al. (2011) Parameterization results in too much OM, here a good agreement with the measurements is found. It is still unclear whether OM acts as additional material to sea salt or replaces it in the emitted particles. The work of Gantt and Meskhidze (2013) found marine aerosols with OM and without sea salt as well as aerosols including both

species. Nevertheless, this is still an open question and therefore modellers have to make clear which approach they use. We extended the part dealing with this topic to clarify this point. In addition we should note that we compared the modelled data to a station in the lower latitudes. The seasonality of OM is less pronounced at this station compared to the mid latitudes of Amsterdam Island and Mace Head.

I recommend the discussion regarding the effect of temperature on the bubble spectra to be removed. Firstly it is quite inaccurate, secondly much detailed discussion can be found elsewhere. The process itself going from ocean bubbles to sea spray size distribution is not as simple as it is proposed here. The radius-depth trajectory (Fig. 35) in Lewis and Schwartz (2004) is for still water at 4 km depth and may not describe accurately bubble population in real environment. See e.g., Fig. 4 in Wu (1981) for water T=14 and 3°C.

- The discussion on the temperature distribution of the bubble spectra was removed.

Most of the figures are also confusing. What does MMS on Fig. 2 stand for? What is the point of Fig. 3? How was Figure 4 generated? Manuscript suggests that the "remaining gaps were filled by linear interpolation." Fig. 2 below shows MODIS Terra and Aqua at 9 km resolution merged data that I was able to download. This figure shows large areas of the open ocean (above 45N) covered by clouds. Current paper does not explain how was [Chl-a] $\sim$ 1 μg/L prescribed to this region. Concerning Figures 15-18, what are the purple and orange boxes depicting? Quantiles? Usually the bar between them represents the mean or median, what is it here if the other colored dashed lines are medians?

- The Figures were improves as follows: We added the explanation of MMS to the caption of Fig. 2. There are two intentions for showing Fig. 3. The main reason is to present the model domain for the "European region". The surface salinity was provided as additional information of a model input field. This parameter is important for the emission of dry sea salt. The low salinity of the Baltic Sea explains the low sodium

concentrations measured at Virolahti II. Regarding Fig. 4: We added the information about the generation of this plot also to the caption. The plot was generated by merging monthly averaged data from MODIS-AQUA and -TERRA. The remaining gaps were filled with the climatological data for the month. In the end some smaller gaps south of 15°N are left. A linear interpolation was done to fill them. It is an artefact due to the interpolation that the region north of 48°N retrieve [chl-a] values of 1g/L, which is unrealistic for that season. We added a comment on that in the revised manuscript. Since with the model assumption that the presence of OM does not change the emitted total PMA the conclusions of this manuscript remain unaffected by the assumption about chlorophyll concentrations at high latitudes. Due to concerns of reviewer 3 we decided to use scatter plots instead of the boxplots in Fig. 15-18.

Abstract, please give the model's name. As it is now, it says "an atmospheric transport model". The abstract should not be vague.

- Model name added

Pg. 379, line 3 – "traces gases" should be "trace gases"

- Corrected

Pg. 380, line 10 – "an" should be "a"

- Left as "an "European" region including Iceland and an "African" region"

Pg. 382, line 8 – "Although many components and chemical species could be found, a large fraction is still unknown" please reword

- Reworded to "A large fraction is still unknown, although many components and chemical species are found in the organic fraction of PMA"

Pg. 391, line 1 – "It is used as driver..."

- Changed to "driver model"

Pg. 392, line 2 – "growth" needs to be "grow"

- Corrected

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