

Response to Referee # 2.

We thank the Referee for useful comments and relevant questions to the work presented. Below, we posted our responses.

In general, the paper is well written and the topic of sufficient interest to warrant publication in ACP, however, the conclusions are thin compared to the number of simulations performed.

Following recommendations of the referee, Conclusions section has been made more solid to highlight the main findings and conclusions resulting from the model tests and the attempts to reconcile calculations with observations. In particular, some of the issues discussed below have been also emphasise the Conclusion section.

Specific comments:

Title: I wouldn't refer to "sea salt "as "pollution".

"Sea salt pollution" has been changed to "sea salt concentrations".

Chapter4 "Measurements": The magnitude of air mass concentrations depends crucially on the upper cut-off and, because sea salt exhibits near ground a strong vertical gradient, on the altitude where measurements are taken. Thus, please, describe the measurement data used for comparison to model data more detailed. The EMEP sampler has no well-defined upper cut-off. Nevertheless, is it possible to give an average percentage of total aerosol mass of particles larger than 10 micron?

The sampling and analysis methods for Na⁺ measurement data used in the paper are described in Section 4, and the list of sites is provided in Table A1. The measurements were taken following recommendations from the EMEP Manual for sampling and chemical analysis (the reference is given). The measurements were performed at a standard height of 2m (or close to it). In the model, 2m concentrations are derived from the concentrations at the model's lowest layer (appr. 45m) applying correction factors. The latter account for the specie's (sea salt particles in this case) dry deposition velocity and are calculated based on an assumption about constant turbulent fluxes in the surface layer.

Concerning the mass of sea salt particles larger than 10 μm captured on the filter pack sampler without an upper cut-off, this information is unfortunately unavailable to us. The only estimate is for Birkenes where simultaneous measurements of aerosols were performed with filter pack and low-volume sampler. These data showed that in total Na⁺(measured with filter pack) the fraction of Na⁺ smaller than 10 μm comprised 90% in June 2006 and 87% in January 2007 (Wenche Aas, NILU, personal communication). It was also observed that during sea salt episodes at coastal and near-coastal sites the coarse fraction was typically enhanced compared to that in long-range transported air masses. Thus, given inevitable uncertainty associated with the upper size limit of model calculated sea salt, we do not think that on average this can be a source of discrepancies between calculated and observed Na⁺ in air. However, larger model underestimations can be expected (and indeed registered) in sea salt episodes. Relevant information and brief discussion has been added in the paper.

Page 161, last par: In order to test the performance of the wet removal scheme, it would be interesting to analyze the agreement between air concentration measurements and model results by separating the days with and without precipitation.

This is an interesting suggestion and we have made some additional work to look separately at the model performance in dry and wet days at individual sites. As comprehensive statistical analysis required development and extension of the existing verification tools, infeasible in such a short time frame, we have looked at daily time-series for Na⁺ in air and precipitation, together with observed precipitation. No clear pattern in model performance between the days with and without precipitation could have been identified, which would indicate local character of the problem. In a

number of cases, the underestimation of Na^+ in precipitation seems to be due to insufficient local wet deposition. However, there are also days when both Na^+ in air and precipitation are predicted well or under/overestimated. It was also found that in many cases (measurement sites, time periods), model underestimation of Na^+ in precipitation was to a large degree due to its severe underestimation of just a few episodes with extremely large (up to 5-10 fold) measured values. Performing a full statistical analysis separating dry and wet days would be very interesting and could give new insights to the problem, therefore we would like to do it in the nearest future. A brief discussion on this issue has been supplemented in the paper.

Chapter 8.2: The horizontal resolution of the two meteorological models, EMEP and SILAM, differ by more than a factor of two. This might influence significantly the wind statistics and subsequently the sea salt source strength and hampers the interpretation of this model comparison. EMEP and SILAM use aerosol schemes of different complexity. Can the authors judge whether the more complex scheme produces results that are more realistic? I miss the results, which justified this chapter.

One of the goals of model inter-comparison exercises is to test the robustness of model results. In this work, an important outcome of comparison of the performance of EMEP and SILAM models against observations (which the paper apparently failed to highlight) is that despite some simplifications and coarser resolution the EMEP model's ability to reproduce sea salt concentrations is comparable (and in some cases better) to the SILAM. However, several cases are discussed in the paper in which the EMEP model performs worse compared to SILAM (e.g. at Finnish sites which are not representative for a $50 \times 50 \text{ km}^2$ grid cell of the EMEP model and which are influenced by the Baltic sea with a rather low salinity).

To elaborate further, EMEP and SILAM models use basically the same parameterisation of sea salt production, but additionally the dependence of sea salt emissions on the water salinity is included in the SILAM. The effect of this dependence is not so important for most of the North Atlantic, but as shown in the paper is quite pronounced for the Baltic Sea. Thus, the main causes for model discrepancies would be the horizontal and vertical resolution and the meteorology used. As the referee correctly pointed out, wind field can differ when calculated at different resolution (which in this work comes in addition to different meteorological drivers used for EMEP and SILAM simulations). In the inter-comparison exercise for EMEP runs we used HIRLAM meteorology, which in fact had been produced on $0.2 \times 0.2^\circ$ resolution. The fields were interpolated to EMEP $50 \times 50 \text{ km}^2$ grid. Verification with surface wind observations at SYNOP sites shows very little difference between the original and interpolated wind speed. Verification of wind speed over seas was not possible though. However, comparison of wind maps on $0.2 \times 0.2^\circ$ and $50 \times 50 \text{ km}^2$ grids shows that the interpolated fields represent fairly well the wind field at the finer resolution, except for some areas with very strong wind gradients associated with atmospheric fronts.

The SILAM model distributes produced sea salt within a thinner lowest layer. Preliminary tests with the EMEP model where three layers with height of 18, 45 and 90m were used show that sea salt concentrations are considerably (up to 50-100%) higher over the seas compared to the standard run with a 90m thick lowest layer. Compared to observations, Na^+ concentrations are somewhat more overestimated at coastal sites, while more underestimated at in-land ones from calculations using thinner lowest layer. On average, the bias goes up from -5% to 24% and the spatial correlation goes down from 0.79 to 0.70 (the results considered are for January 2008).

As to aerosol schemes, neither of these models included aerosol dynamic scheme. The main difference was that SILAM used more size fractions to describe sea salt size distribution. This affects sea salt lifetime mostly through its dry deposition. A series of tests performed recently showed that the resulting sea salt concentrations are quite sensitive to the choice of size distribution and median diameters, especially for coarse particles. However, these uncertainties are considered to be smaller compared to the uncertainties in sea spray production.

Summarising, the comparison between EMEP and SILAM performance showed the consistency of

results (for most of measurement sites) and gave us additional confidence in the soundness of EMEP model sea salt calculations. Therefore we conclude that the overall EMEP model accuracy for sea salt is not significantly corrupted due to a coarse resolution and somewhat simpler sea salt description. Some text has been added in Chapter 8.2 clarifying the purpose and main outcome of the study.

Chapter 9 “Conclusions”: The EMEP model simulates too high air concentrations and too low concentrations in precipitation. The article does not offer a satisfying explanation. Given that the amount of sea salt removed is mainly controlled by the source strength whereas the concentration in air is controlled by the residence time, would it help to enhance both the source strength and the removal rates?

Firstly, we would like to point out that the average model bias for Na⁺ in air is between 0 and 12% for the years with appropriate data coverage, so that “too high air concentrations” sounds somewhat too strong.

As suggested by the referee, extra tests have recently been made in which we increased either sea salt production (SSprod) or 10m wind speed (u10) and at the same time increased sea salt wet scavenging ratio (W). A summary of results is provided here:

		Test 1	Test 2	Test 3	Test 4
		SSprod x 2 W = 2·10 ⁶	SSprod x 1.5 W = 2.5·10 ⁶	u10 x 1.15 W = 2·10 ⁶	u10 x 1.1 W = 2.2·10 ⁶
Na in air	Bias	83%	26%	48%	23%
	R	0.82	0.81	0.82	0.82
Na in prec	Bias	-25%	-39%	-34%	-41%
	R	0.82	0.75	0.78	0.75

The results show that quite considerable (however within reasonable limits) simultaneous increase of sea salt production and wet scavenging has not eliminated underestimation of Na in precipitation, though calculated Na air concentrations get too high. The best results with respect to the average statistics have been achieved in tests 2 and 3. However, these calculations tend to overestimate sea salt at coastal sites, while underestimate it at in-land sites, thus leading to the overestimation of regional gradients. Further work is needed in order to explain the disagreement between calculated and measured wet deposition of sea salt and to reconcile model results with observations.

Some discrepancies might arise from the fact that the measurements have no clear upper cut-off and the vertical resolution is pretty coarse. Vertical soundings of sea salt concentrations in air show a strong vertical gradient near the surface, however, the lowest model grid-box is 90 m thick. Please, comment.

Answers to similar comments are already given above, in relation to questions to Chapter 4 “Measurements”. Comments regarding the flagged issues have been included in conclusions chapter of the paper.

Page 172, the sentences in line 12 “sea spray production could be a factor of 2 greater in surf zone compared to the open ocean” and line 17 “Also, Gong et al. (2002) showed that surf zone sea spray in C’ux was much smaller compared to that for an open ocean” are contradictory.

Page 172, the sentences in line 12: The confusing formulation has been changed to:

“Since the surf zone is rather narrow, its relative area, and thus contribution to the total sea spray,

will be relatively small within the EMEP 50x50 km² grid cell. This is supported by work of Gong et al. (2002), who showed that surf zone sea spray flux was much smaller compared to that for an open ocean and concluded that the surfing contribution to the total sea salt production was negligible on a regional scale.”

All typos pointed out by the reviewer have been corrected.