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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sensitivity of modeled atmospheric nitrogen species to variations in sea salt emissions in the North and Baltic Sea regions" *by* D. Neumann et al.

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Response to the comments of Reviewer #1

We thank the reviewer for the constructive comments on the manuscript. Please find the revised version of the manuscript (acp-2015-758-manuscript-rev01.pdf), the revised version of the manuscript with marked changes (acp-2015-758-manuscript-rev01_diff.pdf), and the revised supplement (acp-2015-758-supplement_text_rev01.docx) in the attached zip file.

0) Rather than a general evaluation of speciated mass concentrations at certain sites, I think that the model evaluation needs to address scientific questions regarding sea salt emissions such as the chemical composition, size distribution, transport/deposition, and interaction with other aerosol components. While some of these evaluations may not be possible with the limited observational dataset, evaluating groups of stations with similar characteristics (inland vs coastal, remote vs urban, agricultural vs industrial, etc.) as opposed to individual stations may help identify strengths/weaknesses of the existing sea spray emission parameterization besides the lack of salinity-dependence.

> We moved the tables showing the statistical comparison at all stations (see item (2)) from the supplement into the manuscript, grouped the stations into coastal and inland stations (> 50 km downwind to the coast), and discuss the differences between these groups. We also removed the stations Schauinsland (DE0003R) and Sniezka (PL0003R) because they are located on mountains in 1205 m and 1603 m height, respectively. We added the stations Melpitz (DE0044R), Utö (FI0009R) and Virolahti II (FI0017R) to have an additional inland and two additional Baltic Sea stations.

> We agree with the reviewer that chemical composition and size distribution of sea salt particles are important parameters in chemistry transport models and need validation. However, we felt that including an analysis of modeled sea salt size distribution would make topics of this manuscript too diverse. Therefore, we submitted another manuscript to ACPD on comparing three different sea salt emission source functions that differ in the considered parameters (wind + salinity vs. wind + SST + salinity vs. wind + SST + salinity + waves) and in the sea salt particle size distribution. Unfortunately, the second manuscript is not available as discussion paper, yet (02-Feb 2).

> An analysis of the nitrogen deposition was added to the results section (new section 3.3), to the discussion section (new section 4.4), to the conclusions (one paragraph) and to the study's objective (a) in the introduction was extended by "*[...] and on nitrogen*

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deposition to the ground and into the sea".

1) Page 29713, Sect. 2.4.3: Please add a more comprehensive description of the salinity inputs to the model.

> The salinity data were taken from an extended version of the HAMSON model. We added references describing the model to Sect. 2.3 and added information on the used data to Sect. 2.4.3.

> Text in Sect. 2.3: "*The salinity data were taken from runs of a hydrodynamic model based on HAMSOM (HAMburg Shelf Ocean Model). A detailed description of the hydrodynamic model and recent updates were published by Schrum and Backhaus (1999) and Barthel et al. (2012), respectively.*"

> Text in Sect. 2.4.3: "*Annual average salinity data from the year 1993 were used. Annual averages were taken because the oceanic data are time independent in CMAQ. Unfortunately, data for the year 2008 were not available to the authors when the CMAQ model runs were performed. According to Matthäus et al. (1997) and Nausch et al. (2009), the difference in the sea surface salinity between the years 1993 and 2008 is low. Its affect on the sea salt emissions is low compared to the difference between salinity-scaled and non-salinity-scaled sea salt emissions (for example, see Fig. S4). Therefore, we assume that employing salinity data from the year 1993 instead of 2008 has no relevant impact on the results of this study.*"

2) Page 29716, Sect. 3.2: It is not clear to me why these three stations were selected for analysis in the main text and the others only in the supplement. As I suggested in my general comments above, grouping of stations with similar characteristics may allow for a more useful evaluation of the model.

> We moved the tables with the statistical data of all stations from the supplement to the

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main manuscript and describe/discuss the data in the text. The stations are grouped into coastal and inland stations, now. A stations is considered to be inland (> 50 km downwind to the coast).

> These three stations at Westerland, Waldhof, and Zingst where chosen to be discussed in detail because they represent North Sea air masses, inland air masses (with coastal influence) and Baltic Sea air masses (with impact from North Sea air masses), respectively (see page 29714, line 12+13). We added the sentence "*Thus, these stations' measurements cover three different air quality regimes.*".

3) Page 29717, Sect. 3.2.2: Despite the fact that the title refers to the sensitivity of nitrogen species to sea salt, the model comparison with sulfate comes before the nitrogen species. I would suggest removing most of the evaluation/discussion of sulfate or change the title to reflect the inclusion of non-nitrogen evaluations.

> We removed the section on corrected sulphate, added a section on nitrogen deposition, and changed the title from "*Sensitivity of modeled atmospheric nitrogen species to variations in sea salt emissions in the North and Baltic Sea regions*" to "*Sensitivity of modeled atmospheric nitrogen species* ***and nitrogen deposition*** *to variations in sea salt emissions in the North and Baltic Sea regions*".

> We removed corrected sulphate and added nitrogen deposition rather than changing the title because – reconsidering the structure of the manuscript – analyzing the impact of sea salt on concentrations of nitrogen species and on nitrogen depositions gives a more complete view. Moreover, analyzing the impact of sea salt particles on non-sea-salt-sulphate from different sectors – e.g. shipping and energy production – in future studies seems more valuable than only analyzing the impact on bulk non-sea-salt-sulphate that is negligible low. Also there are no studies known to us proclaiming opposite results – a strong effect of sea salt particles on non-sea-salt-sulphate.

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4) Page 29717-29718, Sect 3.2.3 and 3.2.4: Comparing the observed and simulated concentrations of nitrogen species based on the sum of the components instead of individually limits the evaluation of the sea salt aerosol chemistry. If these nitrogen components are available individually from the observational dataset, I'd suggest com- paring ammonia, ammonium, nitrate and nitric acid separately.

> We strongly agree with the reviewer that comparing ammonia, ammonium, nitrate, and nitric acid separately would be more meaningful. Unfortunately, the measurements at most EMEP stations in the considered region were performed with 3-filter packs in 2008. Even tough separate measurement values of these four species are available via the EBAS database, they should not be considered individually (page 20714, lines 20 – 23) according to the EMEP manual. Technically, we could compare the separate values but we have no information on the error of the measurements.

> We added a brief evaluation on the impact of sea salt particles on NO_3^- (not sNO_3) at the end of Sect. 3.2.3 and added Fig. 8 and Table 5. It is also discussed in the discussion section.

5) Table 2: Despite sign change in the biases of sodium concentrations at two of the three stations between winter and summer, the text includes little discussion of the seasonal changes besides a general statement of the magnitude. I'd suggest adding Discussion Paperdiscussion of the potential sea surface temperature dependence of the sea salt emissions or other factors which may influence the seasonality.

> We extended the comparison of differences between winter and summer at several locations in the manuscript.

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> We added two paragraphs on the impact of the SST and of the sea surface micro layer (SML) on sea salt emissions in the discussion section:

> "*Based on laboratory studies, Martensson et al. (2003) found that the sea salt emission size spectrum depends on SST. Jaeglé et al. (2011) and Gantt et al. (2015) improved sea salt particle model results by applying SST dependence to sea salt source functions. The results of Martensson et al. (2003), Jaeglé et al. (2011), Callaghan et al. (2014), and Salter et al. (2015) clearly show that sea salt emissions decrease when the SST decreases. The* Na^+ *concentrations might be overestimated at coastal stations during winter because Gong (2003) does not consider the SST when calculating sea salt emissions. However, this factor does not explain the general overestimation in summer.* "

> "*The SML that is formed by mainly surface active organic compounds affects the bubble-bursting process and, thus, sea salt emissions. Because the marine biological activity is higher during summer than during winter, one might expect that the SML affects sea salt emissions more during summer than during winter. This could explain the general overestimation of* Na^+ *concentrations during summer. However, the impact of the SML on sea salt emissions is currently poorly understood and little investigated.* "

> We consider SST dependent sea salt emission parameterizations in another study submitted to ACPD (see reply to (0)).

6) Table 2: The Table 2 caption includes a statistic (NMB) that are not found in the table and vice-versa for RAE

> NMB is removed from the caption and RAE is added.

7) Typos:

7a) Page 29706, line 9: should be "As a model extension"

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7b) Page 29737, figure caption: should be “concentrations”

7c) Pages 29743-29745, figure captions: These figures are not adding species to the graphs but simply replacing them. These captions should remove “the addition of”

> 7a: included

> 7b: included

> 7c: captions modified accordingly

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C12193/2016/acpd-15-C12193-2016-supplement.zip

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