**Atmospheric
Chemistry
and Physics**
Discussions

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Interactive comment on "Sea salt emission, transportation and influence on nitrate simulation: a case study in Europe" *by* Ying Chen et al.

Anonymous Referee #1

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The manuscript *Sea salt emission, transportation and influence on nitrate simulation: a case study in Europe* compares modeling and measurement data obtained during the HOPE-Campaign in September 2013. Sea salt sodium concentrations, nitrate concentrations and the particle size distribution are evaluated at the inland station Melpitz. The concentrations of these species are also evaluated at three coastal Dutch EMEP stations. Moreover, the vertical distribution and the medium range transport of sea salt particles is described and discussed in detail, which is one of the highlight topics of this manuscript. A comparison of modeled columnar particle concentrations with measurements – e.g. via AOD data – would be of great value for this manuscript. The authors employed a coupled meteorology chemistry transport model in this study, which is another highlight. Although a comparison with results obtained via an uncoupled model system would be very interesting, it would be too time consuming to perform such

model runs for this study (maybe the authors could keep this in mind for future studies). However, the authors could highlight the advantages of a coupled model setup in the beginning of the manuscript. The impact of sea salt particles on atmospheric nitrate mass concentrations is analyzed in the end of of the manuscript. The presented and discussed results are not new and could be enhanced or removed (see text to questions 13).

The Figures included in the manuscript as well as in the supplement are of good quality and present the results in a clear manner.

The text is easy to understand but has deficits in the scientific language and in the choice of suitable wording in some passages. Moreover, grammar errors or misspellings complicate the understanding of some long nested sentences. Therefore, a revision of the language is recommended.

1. **Does the paper address relevant scientific questions within the scope of ACP?** Yes. The impact of sea salt particles on other atmospheric compounds and the vertical distribution of sea salt particles were evaluated. Both are topics relevant topics within the scope of ACP.

2. **Does the paper present novel concepts, ideas, tools, or data?** Yes. Most previous model studies on the atmospheric transport of sea salt particles and their impact on other atmospheric compounds were performed with uncoupled meteorology and chemistry transport models. In contrast this study is one of the first evaluating with topic by means of a coupled model system (WRF-Chem). Additionally, the authors evaluate the vertical distribution of sea salt particles. However, the evaluation of atmospheric sea salt concentrations against EMEP measurement data is not novel as well as the evaluation of the impact of sea salt on atmospheric nitrate. The discussion of the vertical sea salt concentration profiles would greatly benefit if measurement data on the column sea salt concentrations were additionally considered - e.g. AERONET AOD data.

3. **Are substantial conclusions reached?** Yes, partly. The authors discussed on a quite detailed level why sea salt particles are transported to a measurement station in the hinterland. Further it is found that sea salt concentrations are over-estimated by the model which a common result of recent model sea salt studies.

4. **Are the scientific methods and assumptions valid and clearly outlined?** Yes, the methods are clearly outlined. The reasoning for some assumptions in section 3.4 should be revised.

5. **Are the results sufficient to support the interpretations and conclusions?** Yes, the results and their representation in the manuscript are sufficient. The re-producibility would be facilitated if the plotted data were attached as supplement information (as text-CSV, netCDF or another appropriate format).

6. **Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?** Yes.

7. **Do the authors give proper credit to related work and clearly indicate their own new/original contribution?** Yes. In a few situations (see my comments below), additional references were appropriate.

8. **Does the title clearly reflect the contents of the paper?** Yes, in principle it does. The authors might consider to add *Northwestern* in front of *Europe* and to replace *simulation* by *in a coupled meteorology and chemistry transport model* (or *in WRF-Chem*). Since the usage of a coupled model is quite novel with re-spect to this topics, it is reasonable to add this information.

9. **Does the abstract provide a concise and complete summary?** Yes, it pro-vides a concise and complete summary and is well written.

10. **Is the overall presentation well structured and clear?** The *Data and Methods*, *Results and Discussions*, and *Conclusions* sections are well structured. Some descriptions in the *Results and Discussions* section should be moved to the *Data and Methods* section as indicated by some of the comments below. The authors might consider to restructure the *Introduction* section. I am missing a clear "story line" in the latter section.

11. **Is the language fluent and precise?** The language is fluent but not completely clear throughout the text. In some situations the used expressions are rather colloquial than scientific. In the comments below, some of these expressions are listed. Although it should be noted that the colloquial expressions make the text easier and more fluent to read than with the correct scientific expression (and partly more lengthly formulations). The usage of the definite article "the" and of the indefinite article ("a" in singular; nothing in plural) is mixed in several passages. In the *Introduction* and section 3.4, some sentence structures are not clear – it is unclear weather spelling or grammar mistakes are the reason. A comma (",") has to be placed after "Thus", "Additionally" and similar words, which start sentences. I suggest to revise the manuscript with a focus on these points.

12. **Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?** Yes. The concentrations of substances are written as [X] and X (where "X" is the substance's chemical formula). The writing should be either [X] or X - not mixed.

13. **Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?** In the current version, section 3.4 does not present new and/or unexpected results. It should be enhanced by new considerations (e.g. the impact of sea salt particles on the vertical distribution of nitrate) or removed.

14. **Are the number and quality of references appropriate?** Yes. Additional refer-

ences might be reasonable in some passages. These passages are listed in the comments below.

15. **Is the amount and quality of supplementary material appropriate?** Yes, the supplements adds value to the main manuscript. I would suggest to put more than one figure/table on each page in order to saves pages and avoid large white spaces. Page numbers would be favorable. Furthermore, the authors should verify whether reprint of the three plots in Fig. 4 violate Copyright laws. Also see the answer to point 5.

General and Scientific Comments:

1. p.1, l.20: "... and has significant impact on the formation on secondary inorganic aerosol particles on global scale.". Reading this sentence might imply that the presence of sea salt particles favors the formation of sec. inorg. aerosols (SIA). However, this is not the case as the authors probably know. Sea salt has an indirect impact on particle formation because compounds such as H2SO4, HNO_3, and HNO_3, which tend to form new particles, condense onto sea salt particles surfaces instead. Hence, the particles formation is decreased. Please reformulate the sentence.

2. p.2, l.8-9: In which context do sea salt particles **participate** in **heterogeneous reactions**? The salt particles provide surface area for heterogeneous reactions but using the work "participate" might be misleading. Please also clarify the meaning of "leading to the formation of secondary aerosols" (see 1.).

3. p.2, l.10: "... significant influence on **nitrate formation** ...". The deprotonation of an acid (HNO_3) should not be denoted as the **formation** of the deprotonated version of this acid (NO_3^-). If nitrate was formed from different compounds via heterogeneous reactions at the particle surface, **nitrate formation** was appropriate. However, the latter situation is not the case, here. The usage of "formation" in connection with "ni-

trate" arises in further text passages, such as p.2, l.17. Please considered replacing "formation".

4. p.2, l.11: "chlorine deficit": Commonly, it is denoted as "chlorine displacement".

5. p.2, l.13-15: Please give the reason for the difference in the cations bound to nitrate between central/western and northern/southern Europe. → extensive animal husbandry in central and western Europe associated with high ammonia emissions.

6. p.2, l.16: "short life-time due to its quick deposition within the coastal region": The other way around: Coarse sea salt particles have a short life time and, therefore, they depose close their source. If the sea salt is emitted close to the coast the it also deposes in coastal regions.

7. p.2, l.24: Please give a reference for the first sentence's statement (and "much more" is colloquial style).

8. p.2, l.25-26: "The parameterization schema . . . ": consider introducing an abbreviation for the schema, such as GO03 as common in the literature.

9. p.2, l.35: Consider starting a new paragraph, here.

10. p.2, l.36-37: "The uncertainty of the SSA emission scheme directly determines the uncertainty of the evaluation of SSE radiative forcing." This is partly correct, because the deposition – particularly variable dry deposition for variable sea salt particle size distributions – plays a relevant role.

11. p.2, l.37-39: Consider switching (and slightly reformulating) the two sentences starting with "Additionally" and "the heterogeneous".

12. p.3, l.3: "nitrate simulation": reformulate; "nitrate prediction"?

13. p.3, l.6-9: Please consider to mention "MOSAIC" and "CBMZ" already in this first paragraph. The detailed explanation further below is fine.

14. p.3, l.6-7: Consider extending ". . . regional air quality model." to ". . . regional mete-orology and air quality model system.".

15. p.3, l.8-9: "In addition to meteorology, aerosols, trace gases and interactive pro-cesses . . . ": "meteorology" was not mentioned before but everything behind meteorol-ogy was indirectly mentioned by writing "air quality model". Please consider reformu-lating the sentence.

16. p.3, l.10-11: Please clarify in the text that MOSAIC is the employed aerosol module in WRF-Chem and not an individual modeling system.

17. p.3, l.13-16: Please state a first, why the bin is split (PM1 and PM1-10 calculation), and then, how it is done.

18. p.3, l.18-19: "Both particle mass concentrations and particle number concentra-tions are simulated.". Question (I am not familiar with the sectional particle representa-tion in MOSAIC): Should the particle number and mass concentrations not be related via the size range of the bin? If the number concentration of particles of a pre-defined size (e.g. 625 nm to 1250 nm in size bin 5) is known, then the particle volume concen-trations (assuming uniform size distribution in this bin) and mass concentrations can be directly calculated. Why are number and mass individually modeled per bin (which could result indirectly in particle sizes outside of the bin's size range).

19. p.3, l.39 to p.4, l.1: Please consider to describe (a) the outer, the intermediate, and the inner domain (in this order) or (b) the inner, intermediate, and outer domain but not (c) the outer, the inner and then the intermediate domain.

20. p.4, l.2: Consider adding "time" or "period" after "spin-up".

21. p.4, l.3: Please add "NCEP" in front of "sea surface temperature". Were the FNL data used as meteorological boundary conditions for the outer model domain?

22. p.4, l.7: Please update the url to MOZART (http://www.acom.ucar.edu/wrf-chem/mozart.shtml).

23. p.4, l.6: "The initial chemical and boundary conditions ...". Please switch the position of "chemical": "The chemical initial and boundary conditions ...".

24. p.4, l.9: Please introduce and describe the F-CASE and and the R-CASE.

25. p.4, l.10: "SSA results from **dried sea spray** ...": The use of "dried" could be misinterpreted by readers as "dry sea salt".

26. p.4, l.12: Neumann et al. (2016) is no primary reference for this statement.

27. p.4, l.20: A side note to the choice of the adjustable parameter: Gantt et al. (2015) (doi: 10.5194/gmd-8-3733-2015) suggests a value of 8 instead of 30.

28. p.5, l.18: "with a temporal resolution of 2 days." The formulation is misleading, because it might be understood by readers that two-day filter samples (48-hour averages) are collected at the Dutch stations. Actually, one-day filter samples (24-hour average) are collected every second day as the authors know and correctly plotted in Fig. 5. Therefore, please reformulate.

29. p.6, l.1: "be unrealistic sources" → "be unrealistic **high** sources"?

30. p.6, l.22-25: The cited studies do not explicitly focus on Northwestern Europe. Tsyro et al. (2011) (doi: 10.5194/acp-11-10367-2011) presented an extensive model study on sea salt concentrations in Europe spanning several years. Manders et al. (2010) (doi: 10.1016/j.atmosenv.2010.03.028) also compared sodium model and measurement data at several EMEP stations. Both found overestimations. Neumann et al. (2016) found overestimations in winter. The authors might consider the refer to these studies, because they focus on a similar region as this manuscript does.

31. p.7, l.3: "...SSA was emitted near the surface layer ...": Sea salt particles should be emitted into the surface apart from the situation, in which sea salt is emitted from the top of a giant wave higher than the model surface layer. However, in the latter situation we probably need another emission parameterization and should not use 10-m wind data.

32. p.7, l.11-22: The authors could consider to describe some information of this paragraph in the Data and Methods section.

33. p.7, l.13: "...promoting the formation of secondary inorganic aerosol (SIA), ...": Secondary aerosols or secondary particles denotes the **new** formation of particles in the atmosphere. The presence sea salt particles enhances the HNO_3/NO_3^- condensation and, hence, one could reformulate the sentence into "...promoting the formation of secondary inorganic particle mass ...".

34. p.7, l.15: "Part of HNO_3 will participate in the partitioning process and form particulate nitrate." Misleading. The whole HNO_3 is involved into the partitioning process. One part remains in the atmosphere and the other part condenses. The condensed part becomes nitrate. → Suggestion: "HNO_3 undergoes (maybe another word) a partitioning process between gas phase and liquid particle phase via condensation. The condensed HNO_3 deprotonates to NO_3^-."

35. p.7, l.18: "irreversible reaction", better "irreversible process"

36. p.7, l.35-36: Why are 5% of the original sea salt emissions emitted in the R-CASE and not 20%, 10%, or 1%? Is this value arbitrarily set or is it related to the 20-fold overestimation of sodium PM10 by the model (1/20 = 5%)?

37. p.7, l.39: "simulation" → "prediction"? (see comment 12.)

38. p.7, l.43-45: Consider switching the order of both sentences.

39. p.8, l.1-7: Data and Methods section?

40. p.8, l.8: "probability density function": "frequency distribution" might be more appropriate (also at the subsequent occurrences in the paragraph)

41. p.8, l.9-10: Please clarify "marine period (Na+ $> 1.8\mu g/m^3$ in F-CASE).". It means that only PFnitrate values of model time steps with [Na+] $> 1.8\mu g/m^3$ were considered? How many model time steps were considered? Please be consistent with the notation

of concentrations: Above, [HNO$_3$] denotes the concentration of HNO$_3$ but, here, Na+ (and not [Na+]) denotes the concentration of Na+.

42. p.8, l.12-13: "also there was uncertainty of the precursors emissions in the model." There are always uncertainties in the emissions. Therefore, the uncertainty is not a general reason not to compare model and measurement data. Note: It should be "precursor's emissions" or "precursor emissions".

43. p.8, l.15: "were used as reference": A reference for what?

44. p.8, l.15-16: " Considering that most of the SSA was **emitted** as coarse mode particles (about 88% in both file measurement and simulation on September 17, 2013 **at Melpitz**), . . . ": The formulation is misleading. There are no emissions measured at Melpitz but concentrations. Since coarse particles have a higher dry deposition velocity than fine particle one can expect that the emissions consisted by more than 88% of coarse particles.

45. p.8, l.17-18: ". . . also more sensitive to the change of the SSA emission.": Why? The particle surface area is the parameter governing the condensation of nitric acid. Higher mass emissions cannot be directly related to higher particle surface area emissions.

46. p.8, l.20-21: ". . . median . . . was about 0.75 . . . broad spreading . . . ": There should be something like ". . . and distribution " between the "0.75" and "broad spreading".

47. p.8, l.20-21: ". . . increased the coarse mode nitrate partitioning fraction by 0.2.": Unclear whether 0.2 is a difference or a quotient. It is the difference, but the formulation is ambiguous.

48. p.8, l.22-24: The conclusion is justified because the ammonium mass concentrations are quite similar in both cases - otherwise not. Therefore, it might be reasonable to repeat that information in this summarizing sentence. Please consider splitting this sentence into two sentences.

49. p.8, l.25-26: "...indicating that in our case the overestimation of SSA emission is mainly in the coarse mode.": What is the reasoning for this conclusion? No comparison against measurement data were performed.

50. p.8, l.28: "consumption of precursors": Why plural? Are there other precursors than HNO_3 for particulate NO_3^-?

51. p.8, l.29: "PM10" → "fine PM" or "PM1"?

52. p.8, l.33; p.9, l.23: "overestimated by 0.2": see comment 46.

53. p.8, l.35: "particle number distribution": There were no number concentrations considered in this study.

54. p.8, l.39; p.9, l.20: "simulation": see comment 12.

55. p.9, l.3, 10, 20, 30: "uncertainty": Uncertainty describes *instable* deviations (in some situations values are overestimated and in other situations they are underestimated). Here, the parameterization clearly overestimates the emissions. Therefore, "uncertainty" is not necessarily the correct word.

56. p.9, l.6: "The variations" → "The spatial variations"

57. p.9, l.8: "...the overestimation in ..." → "...the overestimation of emissions in ..."

58. p.9, l.20: "Fig. 9": Please do not include new Figures in the Conclusions. This Figure should be described in an earlier passage of the manuscript or removed. The first choice is favorable because the figure describes the transport of sea salt particles to Melpitz very well and clear.

59. p.9, l.25: "gas-phase precursors", see comment 50.

60. p.9, l.31: "formation of secondary inorganic aerosol", see comment 33.

61. p.9, l.37-38: Last sentence: If the authors want to write about NO_X it should be

done in an extra paragraph of the *Conclusions* section and not in the last sentence. The second last sentence might be a nice last sentence.

Comments on Figures and Tables:

1. p.16, Table 2: The authors might consider to split the three columns into five (one column each for "Factor" and "R"). "Factor" should be explained in the caption.

2. p.17, Table 3: see comment 61.

3. p.18, Fig. 1: "shown in Figure 5": it is **Figure 6**; "domain 02" == intermediate domain?

4. p.20 and 21, Fig. 3 and 4: The title above the plots is inconsistently written: "10-20th Sep. 2013, Melpitz" (Fig. 3) and "10-20th September, 2013".

5. p.22, Fig. 5: The fact that two y-axes exist per plot should be noted in the caption. The authors could consider to order the plots by the stations' distance to the coast or geographic location (== Melpitz as plot (d)) because it is more intuitive with respect to Fig. 1.

6. p.23, Fig. 6: The authors might consider inverting the x-axis because it is more intuitive for the reader to have the coast on the left and Melpitz on the right.

7. p.24, Fig. 7: The length of the x-axis could be cropped.

8. p.25, Fig. 8: The caption is complicated formulated. Consider reformulating it. Additionally: "probability" (1st and 4th line), see general comment 40.

9. p.26, Fig. 9: see comment 6 to Fig. 6 and general comment 58.

Comments on Language and Spelling:

1. p.1, l.33-36: Please split this sentence into two. schulz 2. p.1, l.41: Please change "Atmospheric aerosol plays" to "Atmospheric aerosols play" to be consistent with the

next sentence ("Further **they** ...").

3. p.1, l.42: Please change "could either be" from conjunctive to indicative ("are either").

4. p.1, l.43: "constitute" → "constituent".

5. p.2, l.3-6: Please reformulate the sentence. There are some typos or the grammar is incorrect.

6. p.2, l.7: no conjunctive, see 3.

7. p.2, l.13: "... sodium nitrate is largely contributed to nitrates ... ": Please use active voice ("... sodium nitrate contributes ... ").

8. p.2, l.16: "quick" → wording

9. p.2, l.16: "...region (Grythe et al., 2014), thus ... " → "...region (Grythe et al., 2014). Thus, ..." (split sentence; comma after "thus").

10. p.2, l.17: "cannot reach the distant inland area." the meaning is clear but colloquial language; Why "**the** distant inland area."?

11. p.2, l.20-21: Please move "later on" to the end of the sentence because it specifies a time.

12. p.2, l.22-23: "...provide an opportunity ... " → colloquial. If the guards in a prison do not look after the prisoners, then they provide an opportunity for a prison break. However, the mechanisms do not provide an opportunity for sea salt particles. Also colloquial: "...make their influence more extensive ... ".

13. p.2, l.26: "...is still highly uncertain ...": please reformulate

14. p.2, l.34: "...in varying degrees ...": possibly "...**by** varying degrees..." might be correct; please check

15. p.2, l.39: "...**needs** the **participation** of ... " → colloquial

C13

16. p.2, l.41-42: "... **make** the importance of SSA indirect effect on nitrate formation over a **broader region**.". Please reformulate.

17. p.2, l.43-44: "In this study ... by a case study ...". Please remove the duplication of "study".

18. p.3, l.21-22: Please reformulate the sentence starting with "However,".

19. p.3, l.23: "The formation mechanism of ...". Please change to "The formation of ..." or "A formation mechanism of ...".

20. p.3, l.38-39: "from September 10-20, 2013". Please change to "from September 10 to 20, 2013" or "in the time period September 10-20, 2013" or choose another formulation.

21. p.3, l.40: "... covers **the** whole Europe, part of the North Sea and the North Africa ...". Please remove the two bold "the" and add an "a" in front of "part" and please do the same in the succeeding lines.

22. p.4, l.11: "... emitted from bubble bursting or breaking waves torn by winds at wave crests." → "... emitted **by** bubble bursting or breaking waves **or** torn **of** wave crests by winds."

23. p.4, l.16: "was" → "is"

24. p.4, l.22: "temporal" → "temporally"

25. p.4, l.24: move "code" behind the bracket ("(SNAP) code")

26. p.4, l.26: insert "the" in front of "anthropogenic emission inventory"

27. p.4, l.31: "consists with" → "has"

28. p.5, l.1-2: "the stations all over ... vertical structures." Please reformulate.

29. p.5, l.42: "bin 05-08" → "bin**s** 05-08". Occurs several times.

30. p.6, l.10-11: Please reformulate the sentence.

31. p.6, l.17: "variance/trend": Maybe "temporal pattern"?

32. p.6, l.36: "400 km away from coast" → "400 km distant to the coast"?

33. p.6, l.39-40: "...about 30-40% of SSA mass concentration was actually transported to the inland (Melpitz) comparing to the coast regions." → "about 30-40% of **the initial** SSA mass **at coastal stations** was actually transported to the inland **station of Melpitz**.".

34. p.6, l.41: "will be discussed": Unclear; Where? When?

35. p.6, l.43-44: "...the warmer sea surface resulted in a higher planetary boundary layer (...) than that over the continent." → "...the warmer sea surface resulted in a higher planetary boundary layer (...) **above the sea than** over the continent."

36. p.7, l.4: "...was able to penetrate ..." → "...penetrated ...".

37. p.7, l.11: "from" → "by"

38. p.7, l.12-14: First part of the sentence unclear. Please reformulate.

39. p.7, l.20: "The participation of SSA ..." → "The presence of SSA ..."

40. p.7, l.25: "Either it could result from inaccurate emission of precursors or an improper chemical pathway." → "The overestimation could result either from inaccurate emissions of precursors or from an improper modeled chemical pathway." (suggestion)

41. p.7, l.26-34: Please reformulate the passage. One can interpret what is meant in this passage but the formulation and sentence structure make the understanding difficult.

42. p.7, l.35: "a sensitive study" → "a sensitivity study"

44. p.9, l.11: "continent" → "the continental"

45. p.9, l.12-15: Split into to sentences and replace "participate" by another verb.

46. p.9, l.17-18: "made the SSA overestimated by a factor of 20 at Melpitz": Consider replacing "made" by "led" or "yielded" and reformulate.

47. p.9, l.20-22: Please reformulate.

48. p.9, l.26-27: "..., resulted from coarse mode nitrate formation with participation of SSA, may slow down the formation of fine mode nitrate." → "..., resulting (or: which resulted) from coarse mode nitrate formation, reduced the formation of fine mode nitrate."

49. p.9, l.32-33: "Later on, these changes will alter ..." → "These changes alter ...".

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-309, 2016.