

Response to comments of referee #2

General Comments:

The manuscript “Sea salt emission, transportation and influence on nitrate simulation: a case study in Europe” studies the transport of sea salt aerosol using the WRF-CHEM model and compares the modelling results to measurements obtained during the HOPE-Campaign in September 2013. The meteorology simulations were validated against surface meteorological observations as well as the vertical distribution of meteorological parameters obtained by radiosonde measurements, and both confirmed that the simulation could capture the meteorological condition very well. The aerosol number/mass concentration distribution, however, displayed a large discrepancy in the coarse mode size range, which the author attributes to overestimated SSA emissions in the model emission scheme. The author studies the difference in thermodynamic stratification over land and sea and points out the mechanism for the long-range transport of SSA, which extends the influencing range of SSA further inland to the Melpitz station. The author further studies the effect of overestimated SSA on particulate nitrate simulation results. Here are some general comments:

Response:

Many thanks to the reviewer for the comments and suggestions. We have improved the manuscript accordingly. The language in the manuscript has also been edited throughout.

The order of Figures was changed in the revised version manuscript. However, in this response we keep the order consistent (unless specified) with the original version manuscript for easily understood. The changes of the Figures order are shown in Table R1.

Table R1. The changing of Figures order in the revised manuscript

<i>Original version</i>	<i>Revised version</i>
<i>Manuscript</i>	
--	<i>Figure 1 (newly added)</i>
<i>Figure 1</i>	<i>Figure 2</i>
<i>Figure 2</i>	<i>Figure 3</i>

<i>Figure 3</i>	<i>Figure 4</i>
<i>Figure 4</i>	<i>Figure 5</i>
--	<i>Figure 6 (newly added)</i>
<i>Figure 5</i>	<i>Figure 7</i>
<i>Figure 6</i>	<i>Figure 8</i>
<i>Figure 7</i>	<i>Figure 9</i>
<i>Figure 8</i>	<i>Figure 10</i>
<i>Figure 9</i>	<i>Figure 11</i>
<i>Supplement</i>	
<i>Figure S1</i>	<i>Figure S1</i>
<i>Figure S2</i>	<i>Replaced by revised version Figure 1</i>
<i>Figure S3</i>	<i>Figure S2</i>
<i>Figure S4</i>	<i>Figure S3</i>
--	<i>Figure S4 (newly added)</i>
<i>Figure S5</i>	<i>Figure S5</i>

(1) The impact of SSA on nitrate partition seems to be nothing new. The author mentions at the end of the conclusions the potential impact of overestimated SSA and nitrate on radiative forcing and aerosol hygroscopicity, it would be perhaps more interesting to see some discussion on that.

Response:

Thank you very much for the comments.

We agree that the influence of sea salt on nitrate has been studied in lots of previous studies (Neumann et al., 2016a; Liu et al., 2015; Im, 2013; Athanasopoulou et al., 2008), but mainly focus on the bulk nitrate mass concentrations and did not show the influence on the nitrate within different size mode (fine mode and coarse mode). In this study, we quantified the sea salt influence on the both fine mode and coarse mode nitrate particles formation respectively.

By looking into size-segregated details, we found that sea salt facilitates the coarse mode nitrate particle (NaNO_3) formation (as found in most previous studies), but it may inhibit the fine mode nitrate particle (NH_4NO_3) formation. This effect can change the particle mass size distribution (PMSD) of nitrate, moves nitrate from fine mode nitrate particles to coarse mode nitrate particles (see Fig. 9), which is crucial for aerosol deposition, hygroscopicity, and optical properties etc. This research could serve as a cornerstone for future detailed research about the impact of sea salt on these properties of nitrate.

However, as pointed out by the reviewers, the re-distribution effect of nitrate PMSD due to the participation of SSA was not clearly highlighted in the manuscript. Therefore, in order to emphasize this scientific point, the title, section 3.4, section of Introduction and Figure 9 have been revised. The detailed revisions are shown as following.

The title has been revised as suggested by reviewer 1#:

“Sea salt emission, transportation and influence on nitrate simulation: a case study in Europe” **changed to**

“Sea salt emission, transport and influence on **size-segregated** nitrate simulation: a case study **in Northwestern Europe by WRF-Chem**”

One paragraph has been added in Section 3.4 in order to clearly show this effect: the influence of SSA on nitrate PMSD, moving nitrate particle from fine mode to coarse mode. As shown below:

“In order to see the influence of SSA on nitrate PMSD in a clearer way, the simulated PMSD during marine period at Melpitz was shown in Fig. 1 (newly added in the revised manuscript). It was clearly shown that the nitrate PMSD decreased in the smaller size bins (bins 01-04) but increased in the larger size bins (bins 05-08). In the F-CASE (Fig. 1b) when the overestimated SSA participated in nitrate particle formation, nitrate particle moved from fine mode to coarse mode compared with the R-CASE (see also Fig. 3).”

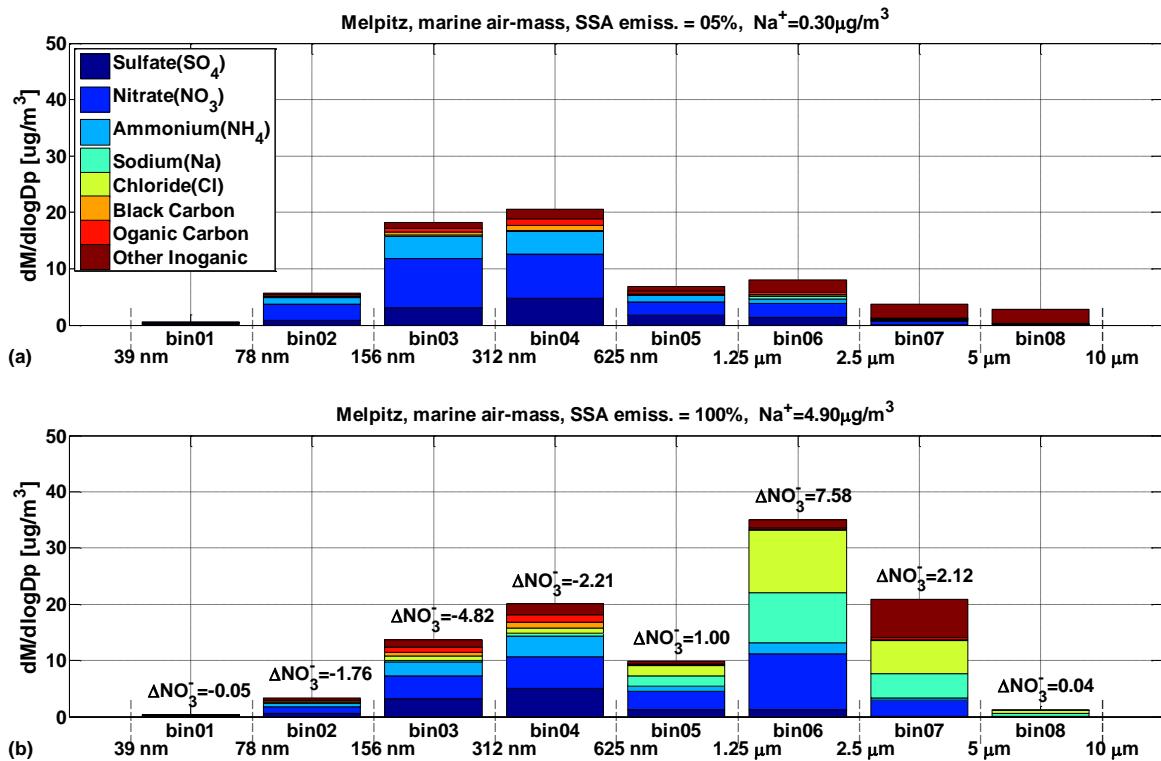


Figure 1 (newly added in the revised manuscript). WRF-Chem simulation results of particle mass size distribution (PMSD) for each chemical compounds, during marine period at Melpitz. (a) result of the R-CASE; (b) result of the F-CASE. The difference of nitrate PMSD between the R-CASE and the F-CASE for each bin is marked.

A paragraph in the Introduction section has been revised, in order to emphasize this scientific point, as shown below:

“SSA could participate in heterogeneous reactions by interacting with trace gases, leading to the formation of secondary aerosols (Seinfeld, 2006), such as nitrate, which is one of the most important secondary inorganic aerosol and is the dominant aerosol component in western and central Europe (Schaap et al., 2011). SSA has a significant influence on nitrate formation as shown in previous studies (Neumann et al., 2016a; Liu et al., 2015; Im, 2013; Athanasopoulou et al., 2008). Sodium nitrate is produced with a chloride deficit in the SSA (Schaap et al., 2011; Seinfeld, 2006), and the timescale of the corresponding reaction is about several hours (Meng and Seinfeld, 1996). As reported in previous studies, sodium nitrate is largely contributed to nitrates in northern and southern Europe (Pakkanen et al., 1999),

whereas in western and central Europe ammonium nitrate dominates (Schaap et al., 2002; ten Brink et al., 1997).” changed to:

“SSA could participate in heterogeneous reactions by interacting with trace gases, leading to the formation of secondary aerosols (Seinfeld, 2006), such as nitrate, which is one of the most important secondary inorganic aerosol and is the dominant aerosol component in western and central Europe (Schaap et al., 2011). SSA can also facilitate the formation of nitrate aerosol (Neumann et al., 2016a; Liu et al., 2015; Im, 2013; Athanasopoulou et al., 2008). However, these previous studies mainly focused on the influence of SSA on bulk nitrate mass concentration, and did not address its influence on size-segregated nitrate particles. In this study, we quantified the SSA influence on both fine mode and coarse mode nitrate particles formation respectively, and the effect could be different for the different size mode, resulting from the heterogeneous reaction on SSA surface with the formation of sodium nitrate. The timescale of this reaction is considered to be several hours (Meng and Seinfeld, 1996). Sodium nitrate is produced with a chloride displacement in the SSA (Schaap et al., 2011; Seinfeld, 2006). Importantly, thermodynamically stable sodium nitrate will not return to the gas phase as the semi-volatile ammonium nitrate does (Schaap et al., 2011). According to previous studies, sodium nitrate largely contributes to nitrates in northern and southern Europe (Pakkanen et al., 1999), whereas in western and central Europe ammonium nitrate dominates (Schaap et al., 2002; ten Brink et al., 1997). The reason is enhanced ammonia emission from husbandry and agricultural sources in central and western Europe (Backes et al., 2016b; Backes et al., 2016a).”

The Figure 9 has been also revised, in order to include this scientific point, as shown below:

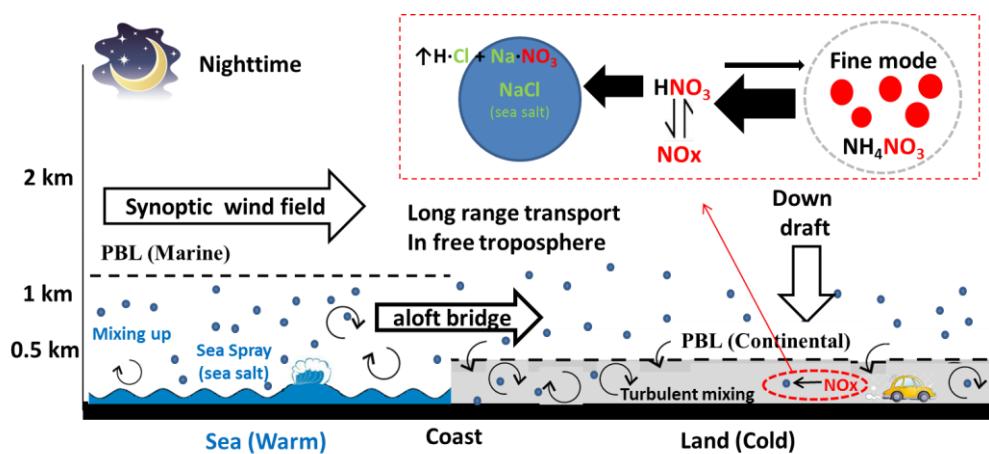


Figure 9. Schematic of sea salt transportation and its influence on nitrate particle formation.

A short discussion of the influence of nitrate PMSD re-distribution on the aerosol particle hygroscopicity, deposition and optical properties has been added in the conclusion, as shown below. The detailed evaluation and studies about these further influences will be presented in the further research paper.

“these changes will alter the physical and chemical aerosol properties, e.g. particle number/mass size distribution and hygroscopicity, which are crucial for climate change evaluation. Furthermore, the direct and indirect radiative forcing evaluation will also be influenced.” changed to:

“Such changes will also alter the physical and chemical aerosol properties, e.g. particle mass size distribution and hygroscopicity. A nitrate coating on a SSA surface may reduce the hygroscopicity of coarse mode particles, and the re-distribution of nitrate from fine mode to coarse mode may increase its deposition rate. Furthermore, the direct and indirect radiative forcing evaluation will also be influenced, since the optical properties (e.g.: single scattering albedo) are strongly related to the size of particles. All these influences are crucial for climate change evaluation.”

(2) The model output frequency is not clarified in section 3. Did you compare hourly model data with observations? In the comparison of simulated & observed meteorological data, the author calculates correlation coefficient. However, many meteorological parameters, such as temperature and wind, have significant diurnal variations, which can be easily captured in the model. If you calculate correlation coefficients between hourly data, the diurnal variations which agree with each other very well might also lead to high correlation coefficients, which does not necessarily mean that you could capture the day-to-day variation well. Why did you not directly compare the absolute values between model & measurements, especially for the wind direction data?

Response:

Thank you very much for the comments. Yes, as reviewer understood, the output frequency is one hour, and the hourly model data was compared with the observations. This has been clarified in the revised manuscript, as shown below.

“*Meteorology simulated by WRF frequency was evaluated with the near-ground measurements at Melpitz and radio-sounding measurements all over Europe.*” **changed to**

“*Meteorology simulated by WRF with hourly output frequency was evaluated with the near-ground measurements at Melpitz and radio-sounding measurements all over Europe.*”

We agree with the reviewer that the agreement of diurnal variations may lead to the high correlation coefficients. And in this study the day-to-day variation was also well captured by the model, as shown in Figure S4. The corresponding sentence has also been revised, as shown below.

“*Simulated temperature, relative humidity, wind speed and wind direction were in good agreement with measurements, with a correlation coefficients (R) of 0.94, 0.85, 0.86, and 0.86 respectively.*” **changed to:**

“*Simulated temperature, relative humidity, wind speed and wind direction were in good agreement with Melpitz near-ground hourly measurements (Fig. S4, newly added in the revised version), with a correlation coefficients (R) of 0.94, 0.85, 0.86 and 0.86 respectively, and with mean bias (MB) 0.38 °C, 9.1%, -0.18 m s⁻¹ and 10.62° respectively.*”

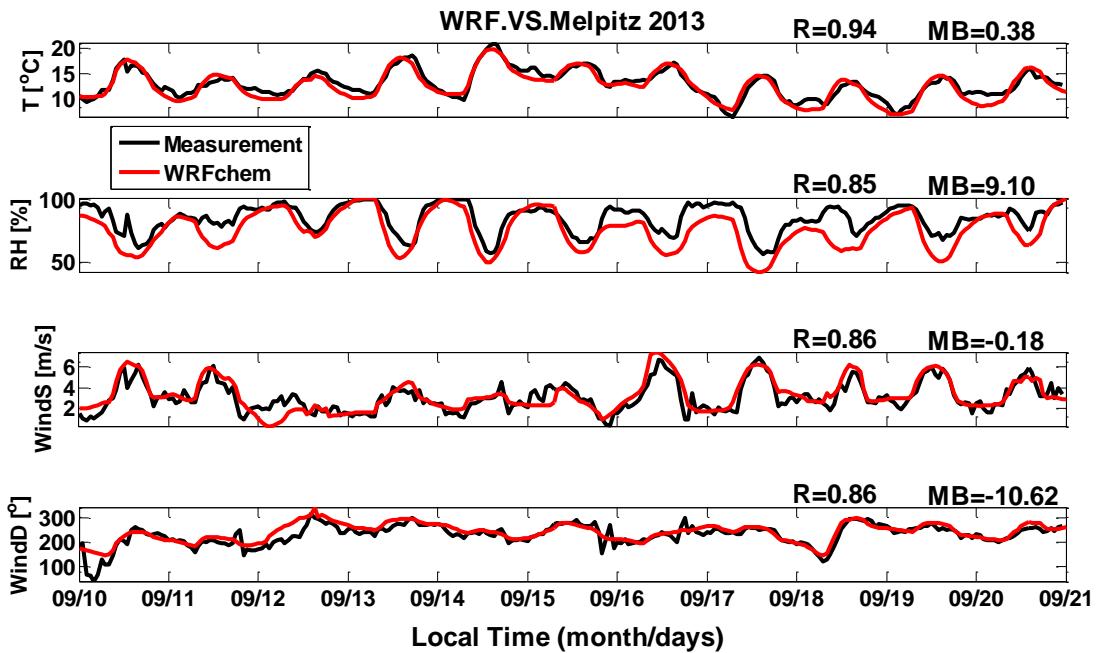


Figure S4 (newly added in the revised version). The comparisons between the simulation results and measurements at Melpitz near-ground layer. The correlation coefficient (R) and mean bias (MB) are marked on the top of each panel. (a) Temperature; (b) relative humidity

(RH); (c) wind speed; (d) wind direction.

(3) Although the manuscript is easy to understand, there are still many grammatical errors and the scientific language is not always precise, please go through the whole text carefully and revise the language to improve the reading experience of your readers.

Response:

Thank you very much for the comments. The language has been edited throughout.

(4) 1. Does the paper address relevant scientific questions within the scope of ACP?

Yes.

2. Does the paper present novel concepts, ideas, tools, or data?

Yes.

3. Are substantial conclusions reached?

Yes.

4. Are the scientific methods and assumptions valid and clearly outlined?

Yes. However, there can be improvements in the methods section.

5. Are the results sufficient to support the interpretations and conclusions?

Yes.

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.

8. Does the title clearly reflect the contents of the paper?

Yes.

9. Does the abstract provide a concise and complete summary?

Yes.

10. Is the overall presentation well structured and clear?

Yes.

11. Is the language fluent and precise?

It is overall fluent, however, improvements are needed to make it more precise.

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

Yes.

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

No.

14. Are the number and quality of references appropriate?

Yes.

15. Is the amount and quality of supplementary material appropriate?

Yes.

Response:

Thank you very much for the comments. The method section has been improved. The language has also been edited throughout.

Specific Comments: Abstract

(1) P1L26: “..., the modeled SSA concentrations were overestimated by a factor of 8-20.” → “, the model overestimated SSA concentrations by factors of 8-20.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(2) P1L27: “...over North Sea” → “...over **the** North Sea”, this needs also to be corrected for the later occurrences in the manuscript.

Response:

Thank you very much for the comments. The terminology has been corrected as suggested.

(3) P1L32: “broadened” → “extended”

Response:

Thank you very much for the comments. The word has been corrected as suggested.

(4) P1L35-36: “increased by about 0.2 for the coarse mode nitrate...., but no significant difference in the partitioning fraction for the fine mode nitrate.” → “increased by about **20%** for the coarse mode nitrate..., but no significant difference in the partitioning fraction for the fine mode nitrate **was found**.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

Specific Comments: Introduction

(1) P1L41: “Atmospheric aerosol plays... Further they have an ...” rephrase these two sentences, if you want to use “they”, you should change the first sentence to “Atmospheric aerosols...”

Response:

Thank you very much for the comments. The “Atmospheric aerosol...” has been revised to “Atmospheric aerosols...”.

(2) P1L43: change to “on a global scale”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(3) P2L1: “..., possibly comparable with...”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(4) P2L3-5: Rephrase to “Waves breaking in the surf zone, where **there are** more whitecaps and stronger **SSA (?)** emission due to **increased** ocean bottom and higher intensity of wave breaking, may **affect SSA concentrations at areas within 25 km distance from** the coastline and can dominate **the SSA concentration** at the coastal region”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(5) P2L9-10: “nitrate formation” is slightly inappropriate, since the HNO₃ was already formed in the atmosphere. The SSA only influenced its gas and aerosol phase partitioning. Please consider

Response:

Thank you very much for the comments. The terminology has been revised to “nitrate particle formation”.

(6) P2L13-14: Change to “..., sodium nitrate largely contributes to nitrates in northern and southern Europe”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(7) P2L22-23: Change to “...and thereby could expand/extend their influencing range from coastal to regional or even global.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(8) P2L24-25: Change to “However, in terms of global mass concentration, ...”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(9) P2L35: Change to “...for the evaluation of its climate effect”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(10) P2L41-42: Change to “Furthermore, the long-range transport mechanisms, as mentioned above, extends the impact of SSA indirect effect on nitrate formation to a broader region.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(11) P2L44: Rephrase as “The model parameterization schemes...”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(12) P3L1-3: Please change the tense in these three lines to present tense.

Response:

Thank you very much for the comments. The sentences have been revised as suggested.

Specific Comments: Section 2

(1) P3L41: Consider adding the domain range of D01 to Figure 1.

Response:

Thank you very much for the comments. In order to see more detail of our interesting region, we prefer to just show D02 in Figure 1, instead of imbedding D02 inside D01 in Figure 1. However, the range of D01 and its relative location with D02 are given in Chen et al. (2016). We have added this information in the manuscript:

“More details on simulation about setups and parameterizations are given in Table 1 and Chen et al. (2016).”

(2) P4L2: “The spin-up **time** of the model run was 2 days.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(3) P4L8: “More details **on simulation** about setups and parameterizations ~~of the simulation~~ are given in Table 1.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(4) P4L10: Rephrase to “SSA are produced through the evaporation of sea sprays, which were ejected into the atmosphere from the sea surface.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(5) P4L12-13: “The parameterization scheme for SSA emission coupled in the WRF-Chem model follows the Gong (2003) scheme.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(6) P4L17: “..., which controls the shape of submicron **SSA** size distributions”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(7) P4L31: “...and has ~~e~~onists with the same spatial resolution”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(8) P4L42: “Measurements of **the** HOPE-Campaign”. The “the” is often missing, please go through the manuscript carefully and make the language more fluent.

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(9) P5L3: “The Melpitz Observatory is representative of for the regional background of Central Europe”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(10) P5L5,9: There are many abbreviations in the text that appear without explaining what they stand for, e.g. WMO-GAW, ACTRIS, MARGA, etc.

Response:

Thank you very much for the comments. The full names of the abbreviations have been added.

As shown below:

WMO-GAW (World Meteorological Organization – Global Atmospheric Watch);

ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network);

MARGA (Monitor for AeRosols and GAses in ambient air).

(11) P5L11-12: “This instrument provided 1-hour data of secondary inorganic aerosols (...) and

gaseous counterparts (...).” I would suggest adding the detailed species that were measured into these brackets.

Response:

Thank you very much for the comments. The detailed species have been added. As shown below:

“secondary inorganic aerosols (NH_4^+ , NO_3^- , SO_4^{2-} , Cl^- , Ca^{2+} , Mg^{2+} and K^+) and gaseous counterparts (NH_3 , HNO_3 , HNO_2 , SO_2 , HCl). ”

(12) P5L12-14: Did you have two high volume samplers respectively for PM10 and PM1? If yes, rephrase to: “The high volume samplers DIGITEL DHA-80 (Walter Riemer Messtechnik, Germany), with a sampling flow of about 30 m³h⁻¹, **were used to collect 24-hour PM10 and PM1 filter samples simultaneously** (Spindler et al., 2013).

Response:

Thank you very much for the comments. Yes, we have two high volume samplers. Therefore we use the proposed text.

(13) P5L14-16: “Information on the **coarse mode (PM1-10)** aerosol chemical compositions, such as nitrate and sodium etc., ~~in the coarse mode (PM1-10)~~ were obtained from the difference between the results of PM10 and PM1”.

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(14) P5L14-16: “Additionally, **24-hour** filter sampler measurements with PM10 inlets (EMEP, 2014) at 3 coastal EMEP station near the SSA transportation pathway (Bilthoven, Vredepeel, and Kollumerwaad, see Fig. 1), **which were collected every second day**, were obtained from EBAS (<http://ebas.nilu.no/>)”

Response:

Thank you very much for the comments. We wrote the sentence as proposed.

Specific Comments: Section 3

(1) P5L21: “over ~~the~~ Northern Germany”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(2) P5L25: “Evidently, strong vertical **motion** occurred in the coastal region, which **resulted in** lifted SSA upward.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(3) P5L28-29: “Simulated **surface** temperature, relative humidity, wind speed and wind direction were in good agreement with **ground** measurements, with a—correlation coefficients...”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(4) P5L36-37: “Corresponding, R values were 0.99, 0.96, 0.84 and 0.92 for potential temperature, wind speed, wind direction and water vapor mixing ratio, respectively.” Are these vertically averaged correlation coefficients between simulated vertical profiles and radiosonde measurements? If so, please rephrase the sentence to make that clear.

Response:

Thank you very much for the comments. The sentence has been revised to make it clearer. As shown below:

“Corresponding, the averaged R values of vertical profiles were 0.99, 0.96, 0.84 and 0.92 for potential temperature, wind speed, wind direction and water vapor mixing ratio, respectively.”

(5) P6L1: Rephrase as “Therefore, unrealistic sources of coarse particles might be the cause for the overestimation.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(6) P6L13-14: “Marine air masses **first** arrived at **the** three coastal stations.

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(7) P6L17: “As shown in Fig. 5 the **day-to-day variation** of Na⁺ concentrations can be captured by the model...”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(8) P6L26-27: “The uncertainties of this scheme may be **attributed** to **the lack** of parameters, ...”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(9) P6L32-33: “Generally, SSA is mostly in coarse mode with **a** lifetime shorter than 2 days in the continental boundary layer, **whereas and reaching** about 1 week in free troposphere”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(10) P6L35: This sentence is hard to understand and needs rephrasing, consider “According to the simulation results, the component of the 10m wind vector that is directed from the coast to Melpitz shows a wind speed in the range of $2-3 \text{ m s}^{-1}$ ”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(11) P6L35-36: “It **would** ~~took~~ therefore **take** about 1.5-2 days for SSA to be transported to Melpitz ($\sim 400 \text{ km}$ away from coast).”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(12) P6L36-38: The result (Fig. S5) from **the** Deposition-Lifetime Concept Model (Chen et al., 2016; Croft et al., 2014) **indicates** that on average only ~~about~~ 10-35% of **the emitted** SSA could be transported to Melpitz through the surface pathway.

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(13) P7L6-8: “Therefore, about 70-85% of SSA (Fig. S5) could be carried further towards **the** inland in free troposphere, and arrived **at** the Melpitz region in the early morning of September 17 (Fig. 6b).”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(14) P7L11-12: “As discussed above, the **over-production** of SSA from the WRF-Chem SSA emission scheme will **lead** to an 8-20 times overestimation of the primary sea salt mass concentration.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(15) P7L15: Rephrase to: “Part of HNO₃ will be partitioned into the condensed phase and form particulate nitrate.”

Response:

Thank you very much for the comments. The sentence has been revised by the reviewer 1#. As shown below:

“The condensed HNO₃ deprotonates to NO₃⁻”.

(16) P7L17-18: “**Another**–The other one is the irreversibly reaction with SSA (NaCl) and the formation of sodium nitrate with depletion of chloride.

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(17) P7L21-22: I believe what you want to say is that the condensation process of HNO₃ onto particles is facilitated by the participation of SSA, replace “partition” with “condensation”: “The participation of SSA might facilitate the **condensation** process of nitrate.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(18) P7L25: “This could either result from an inaccurate emission of precursors or from an improper chemical pathway in the model.”

Response:

Thank you very much for the comments. This sentence has been removed as suggested by the reviewer 3#.

(19) P7L30-34: Please consider rephrasing this part into: “~~The difference between Fig. 7a and Fig. 7b indicates that~~, However, **even under the same mass concentrations of precursors, the simulated nitrate mass concentrations (Fig. 7a) were still much higher than the observed ones (Fig. 7b), which indicates that** in addition to an overestimation caused by overestimated NH₃ emission (see also Table 2), improper chemical pathway also contributed to the nitrate overestimation. ~~Since the simulated nitrate mass concentrations (Fig. 7a) were still much higher than the observed one (Fig. 7b), even under the same mass concentrations of precursors.~~”

Response:

Thank you very much for the comments. The sentences have been revised as suggested.

(20) P7L35-36: “In order to quantify the influence of NaCl on the nitrate partitioning, a **sensitivity** study was implemented with only 5% of SSA emission (R-CASE).”

Response:

Thank you very much for the comments. The sentence has been revised as suggested. And according to the suggestion of the reviewer 1#, this sentence has been moved to the section 2 in order to introduce the R-CASE in the method section.

(21) P7L42: “However, NOx and total ammonia **concentration** results of **the R-CASE did not show significant changes** (Table 2).”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(22) P8L10-13: 1. The later sentence is incomplete; 2. The difference in size range is a reasonable reason why the two should not be directly compared. The uncertainties in measurements and in the model emissions always exist, we need to keep those in mind when comparing measurements with model results, but they are not the reason why the two should not be compared. Consider rephrasing this part into: “Since the MARGA measurements were only available for the size range of PM10, PF_nitrate derived from MARGA observations should not be directly compared with the simulated one. Additionally, we need to keep in mind that high uncertainties exist in the HNO3 measurements due to its sticky property and in the model precursor emissions, which brings further difficulty into the comparison between measurements and simulation.”

Response:

Thank you very much for the comments. The sentences have been revised as suggested.

(23) P8L18-20: This sentence needs rephrasing, consider “As shown in Fig. 8a and Fig. 8b, the median value of coarse mode PF_nitrate in the R-CASE was about 0.75, with the distribution broadly spread in the range of ~0.2 to 1, whereas in the F-CASE the median value increased to 0.96, with a much narrower distribution.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(24) P8L26-27: “Although the fine mode PF_nitrate **revealed** no significant difference between R-CASE and F-CASE simulations...”

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

Specific Comments: Conclusion

(1) P8L39-40: "..., the WRF-Chem model was used to simulate **the aerosol** physical and chemical properties during **the** HOPE Campaign..."

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(2) P9L2-4: The overestimate in coarse mode nitrate is also caused by the overestimate in SSA emissions, which is also summarized later on in the following text. I would suggest not to mention it here, rephrase as: "The coarse mode particles were, however, significantly overestimated both in number and mass, due to an overestimate in SSA emissions caused by the current SSA emission scheme.

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(3) P9L6: "The **day-to-day** variations of SSA mass concentrations..."

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(4) P9L19-20: Change to "The overestimation in SSA emissions not only influences the primary SSA simulation itself, but also leads to significant uncertainties in the particulate nitrate simulation."

Response:

Thank you very much for the comments. The sentence has been revised as suggested.

(5) P9L25: “However, the increased consumption of the gas-phase precursor (HNO₃), caused by the coarse mode nitrate formation with the participation of SSA, may inhibit/repress/reduce (?) the formation of fine mode nitrate.”

Response:

Thank you very much for the comments. The sentence has been revised as suggested. As shown below:

“The nitrate partitioning fraction of fine mode was insensitive to the SSA emission. However, the increased consumption of the gas-phase precursor, caused by the coarse mode nitrate formation with the participation of SSA, may reduce the formation of fine mode nitrate.”

(6) P9L35-39: Change to: “Due to the “aloft bridge” transport mechanism, as described in this paper, the influences of SSA are not only confined to the coastal region, but are extended to a broader region reaching as far as 400 km from coast. Meanwhile, the outflow of continental air mass can transport NO_x to the ocean region (Fig. S1), where these influences of SSA on nitrate may also be significant.”

Response:

Thank you very much for the comments. The sentences have been revised as suggested.

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