

## **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:

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.
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?
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.

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.

### **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.
2. P1L27: "...over North Sea..." → "...over **the** North Sea...", this needs also to be corrected for the later occurrences in the manuscript.
3. P1L32: "broadened" → "extended"
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.**"

#### **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..."
2. P1L43: change to "on **a** global scale"
3. P2L1: "..., ~~possibly~~ comparable with..."
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"
5. P2L9-10: "nitrate formation" is slightly inappropriate, since the HNO<sub>3</sub> was already formed in the atmosphere. The SSA only influenced its gas and aerosol phase partitioning. Please consider

rephrasing.

6. P2L13-14: Change to “..., sodium nitrate largely contributes to nitrates in northern and southern Europe”
7. P2L22-23: Change to “...and thereby could expand/extend their influencing range from coastal to regional or even global.”
8. P2L24-25: Change to “However, in terms of global mass concentration, ...”
9. P2L35: Change to “...for the evaluation of the **its** climate effect”
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.”
11. P2L44: Rephrase as “The model parameterization schemes...”
12. P3L1-3: Please change the tense in these three lines to present tense.

## **Section 2**

1. P3L41: Consider adding the domain range of D01 to Figure 1.
2. P4L2: “The spin-up **time** of the model run was 2 days.”
3. P4L8: “More details **on simulation** ~~about~~ setups and parameterizations ~~of the simulation~~ are given in Table 1.”
4. P4L10: Rephrase to “SSA are produced through the evaporation of sea sprays, which were ejected into the atmosphere from the sea surface.”
5. P4L12-13: “The parameterization scheme for SSA emission coupled in the WRF-Chem model follows the Gong (2003) scheme.”
6. P4L17: “..., which controls the shape of submicron **SSA** size distributions”
7. P4L31: “...and has ~~consists with~~ the same spatial resolution”
8. P4L42: “Measurements of **the** HOPE-Campaign”. The “the” is often missing, please go through the manuscript carefully and make the language more fluent.
9. P5L3: “The Melpitz Observatory is representative of ~~for~~ the regional background of Central Europe”
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.
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.

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 Rieme Messtechnik, Germany), with a sampling flow of about  $30 \text{ m}^3\text{h}^{-1}$ , **were used to collect 24-hour PM10 and PM1 filter samples simultaneously** (Spindler et al., 2013).
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.
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 Kollumerwaard, see Fig. 1), **which were collected every second day**, were obtained from EBAS (<http://ebas.nilu.no/>)”

### Section 3

1. P5L21: “over ~~the~~ Northern Germany”
2. P5L25: “Evidently, strong vertical **motion** occurred in the coastal region, which ~~resulted in~~ lifted SSA upward.”
3. P5L28-29: “Simulated **surface** temperature, relative humidity, wind speed and wind direction were in good agreement with **ground** measurements, with ~~a~~ correlation coefficients...”
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.
5. P6L1: Rephrase as “Therefore, unrealistic sources of coarse particles might be the cause for the overestimation.”
6. P6L13-14: “Marine air masses **first** arrived at **the** three coastal stations.
7. P6L17: “As shown in Fig. 5 the **day-to-day variation** of  $\text{Na}^+$  concentrations can be captured by the model...”

8. P6L26-27: “The uncertainties of this scheme may be **attributed to the lack** of parameters, ...”
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”
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 m s<sup>-1</sup>”
11. P6L35-36 : “It ~~would take~~ therefore **take** about 1.5-2 days for SSA to be transported to Melpitz (~400 km away from coast).”
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.
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).”
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.”
15. P7L15: Rephrase to: “Part of HNO<sub>3</sub> will be partitioned into the condensed phase and form particulate nitrate.”
16. P7L17-18: “~~Another~~ The other one is the irreversibly reaction with SSA (NaCl) and the formation of sodium nitrate with depletion of chloride.
17. P7L21-22: I believe what you want to say is that the condensation process of HNO<sub>3</sub> onto particles is facilitated by the participation of SSA, replace “partition” with “condensation”: “The participation of SSA might facilitate the **condensation** process of nitrate.”
18. P7L25: “This could either result from an inaccurate emission of precursors or from an improper chemical pathway in the model.”
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

$\text{NH}_3$  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.”~~

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).”
21. P7L42: “However, NO<sub>x</sub> and total ammonia **concentration** results of **the R-CASE did not show significant changes** (Table 2).”
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 PM<sub>10</sub>, 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 HNO<sub>3</sub> measurements due to its sticky property and in the model precursor emissions, which brings further difficulty into the comparison between measurements and simulation.”
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.”
24. P8L26-27: “Although the fine mode PF\_nitrate **revealed** no significant difference between R-CASE and F-CASE simulations...”

## Conclusions

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

3. P9L6: “The **day-to-day** variations of SSA mass concentrations...”
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.”
5. P9L25: “However, the increased consumption of the gas-phase precursor ( $\text{HNO}_3$ ), **caused by the** coarse mode nitrate formation with **the** participation of SSA, may **inhibit/repress/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  $\text{NO}_x$  to the ocean region (Fig. S1), where these influences of SSA on nitrate may also be significant.”