Response to comments of referee #3

General Comments:

The authors apply WRF-Chem to investigate the effect of sea salt on aerosol nitrate concentrations and the transport mechanisms of sea salt aerosol to an inland site. Additionally, the results of the applied WRF-Chem setup are evaluated against observations. Although the impact of sea salt on aerosol nitrate in general is nothing new, the paper includes sufficient novel aspects and interesting details for a publication in ACP. One important finding is the overestimation of sea salt emissions by WRF-Chem's Gong (2003) sea salt emission scheme. Some more in-depth discussion seems desirable here, e.g. how well the wind speed in the source area are represented or how the applied scheme compares against the other sea salt emission schemes which are included in WRF-Chem. The paper is easily comprehensible. However, it includes numerous language lapses, such as wrong usage of singular and plural, missing articles etc. The co-authors are requested to support the lead author here. Also, some of the figures could be improved in some aspects.

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 Figures order are shown in Table R1.

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

Original version	***Revised version***
Manuscript	
--	*Figure 1 (newly added)*
Figure 1	*Figure 2*
Figure 2	*Figure 3*
Figure 3	*Figure 4*

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

(1) Although the impact of sea salt on aerosol nitrate in general is nothing new

Response:

Thank you very much for the comments.

We agree that the influence of sea salt on nitrate has been studied in 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 shown 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 highlight out 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)."

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 can 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)."

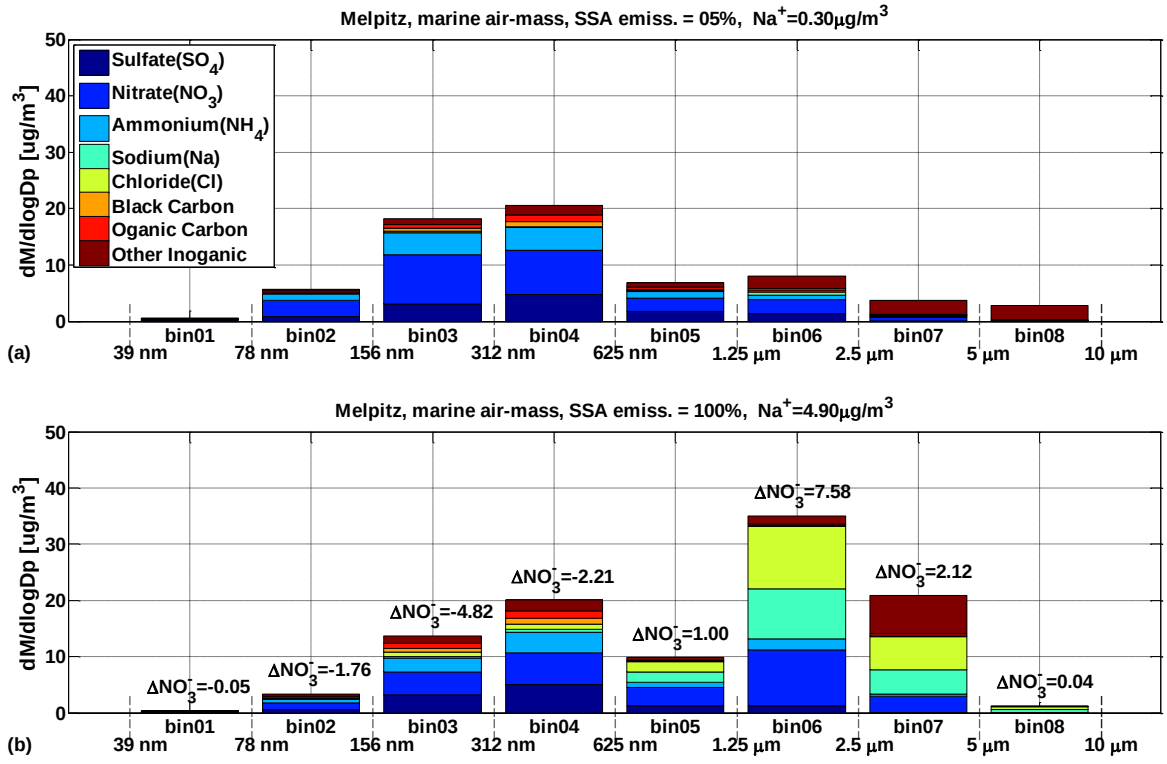


Figure 1 (newly added in the revised manuscript). *WRF-Chem simulation results of particle mass size distribution (PMSD) for each chemical compounds, during marine period at Melpitz. (a) result of the R-CASE; (b) result of the F-CASE. The difference of nitrate PMSD between the R-CASE and the F-CASE for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

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

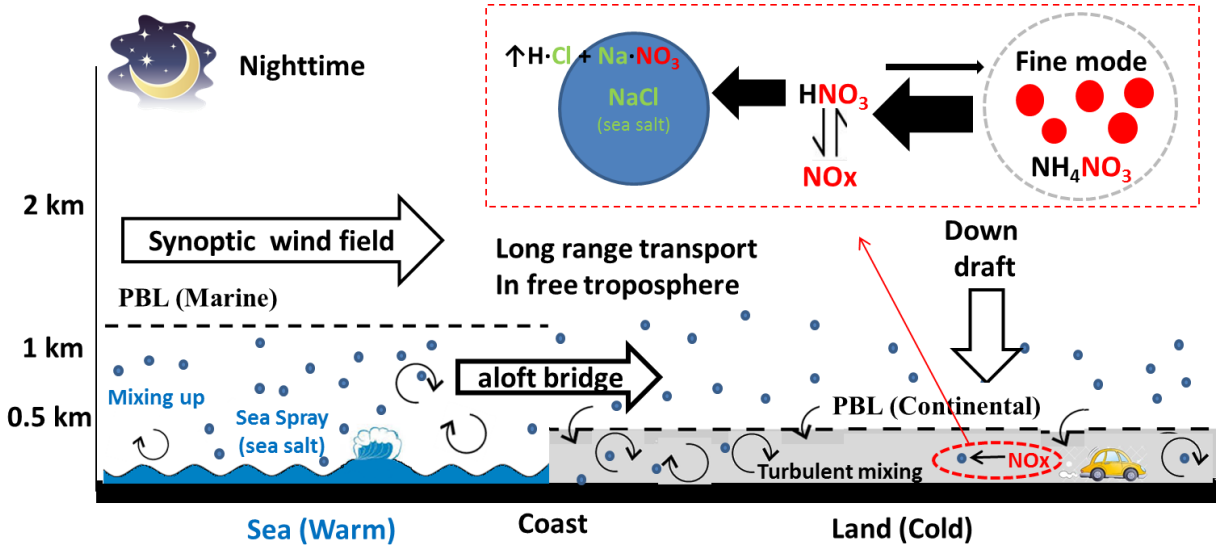


Figure 9. *Schematic of sea salt transportation and its influence on nitrate particle formation.*

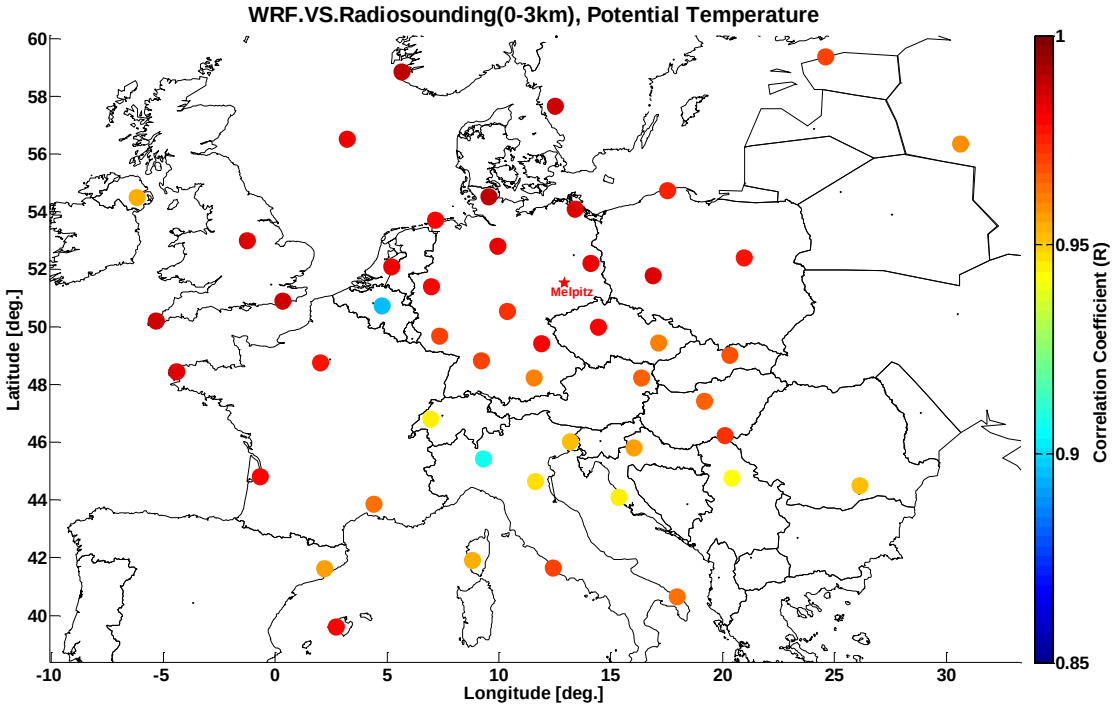
(2) how well the wind speed in the source area are represented

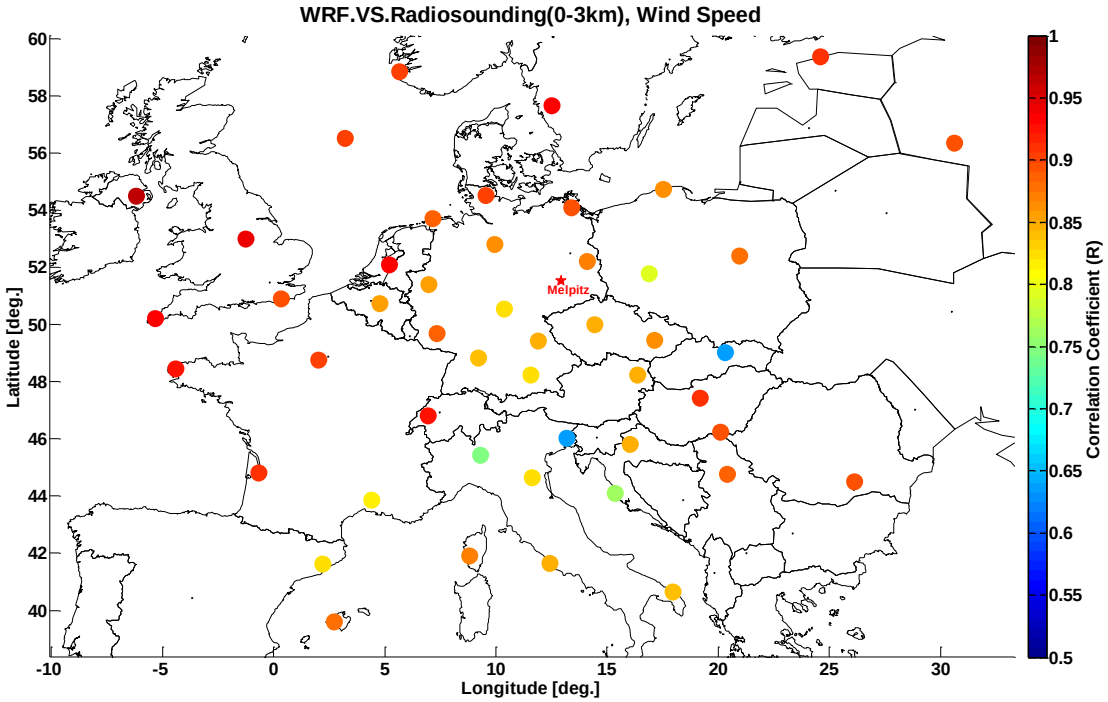
Response:

The ground wind measurements in the source area (over the North Sea) are not available. However, we compared the radio-sounding measurements of wind speed over the Europe, including one station over the North Sea and some coastal stations, which are close to the source area. As shown in Fig. 2b (newly added in the revised version). And the following paragraph has been added in the section 3.1 to discuss about the wind speed simulation.

"The vertical pattern of wind speed was also captured by the model, especially well captured over the North Sea and coastal regions (see Fig. 2b). Generally, the correlation coefficient (R) values were higher than 0.6, with the value higher than 0.9 over the SSA emission source area (the North Sea) and coastal regions."

(a)





(b)

Figure 2. *Correlation coefficient (R) map between WRF-Chem model and radio-sounding measurements (0-3 km). Melpitz is marked as red star. (a) potential temperature; (b) wind speed. Note that the panels (a) and (b) have the different color-bar scale in order to show more details.*

(3) how the applied scheme compares against the other sea salt emission schemes which are included in WRF-Chem.

Response:

Thank you very much for the comments. Unfortunately, there is only Gong (2003) sea salt emission scheme (GO03) is included in WRF-Chem currently. And this scheme is not only coupled with the regional model WRF-Chem, but also with some global models such as GEOS-Chem. Therefore, we think it is important to point out the uncertainties of this GO03 scheme.

Jaegl é et al. (2011) also reported that GO03 overestimated coarse mode sea salt by a factor of 2–3 at high wind speeds over the cold waters of the South Pacific, North Pacific and North Atlantic Oceans, by GEOS-Chem model. The comparisons of GO03 with other sea salt emission functions have also been discussed in Jaegl é et al. (2011). The other emission functions include: (1) adjustment with quadratic wind speed dependence; (2) adjustment with

sea surface temperature dependence; (3) Monahan et al. (1986) scheme; and (4) Clarke et al. (2006) scheme; (5) Mårtensson et al. (2003) scheme. However we should keep in mind that, with the coarse spatial resolution in global model simulation, the detailed PBL structure cannot be properly captured. And this "aloft bridge" transport mechanism, which reported in our research, may be neglected by the global model. Also, the chemical and physical properties of size-resolved aerosol particles cannot be represented in detail by the global model simulation. So, the impact of sea salt on nitrate PMSD was not investigated in the global model studies. In this paper, we would like to investigate the impact of sea salt on nitrate PMSD. And we also would like to introduce a long-range transport mechanism which could expand this impact to the further inland regions, instead of be confined to the coastal regions.

Detailed Comments:

(1) P 5, l 30: How were the correlations calculated, from hourly values or from mean values? How well are spatial patterns represented? Please discuss also absolute error or mean bias.

Response:

Thanks for the comments. The corresponding discussion has been revised, and as suggested by reviewer 2# a new figure (Figure S4, newly added in the revised version) has been added. And the discussion 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 ^{o}C, 9.1%, -0.18 m s^{-1} and 10.62^{o} respectively."

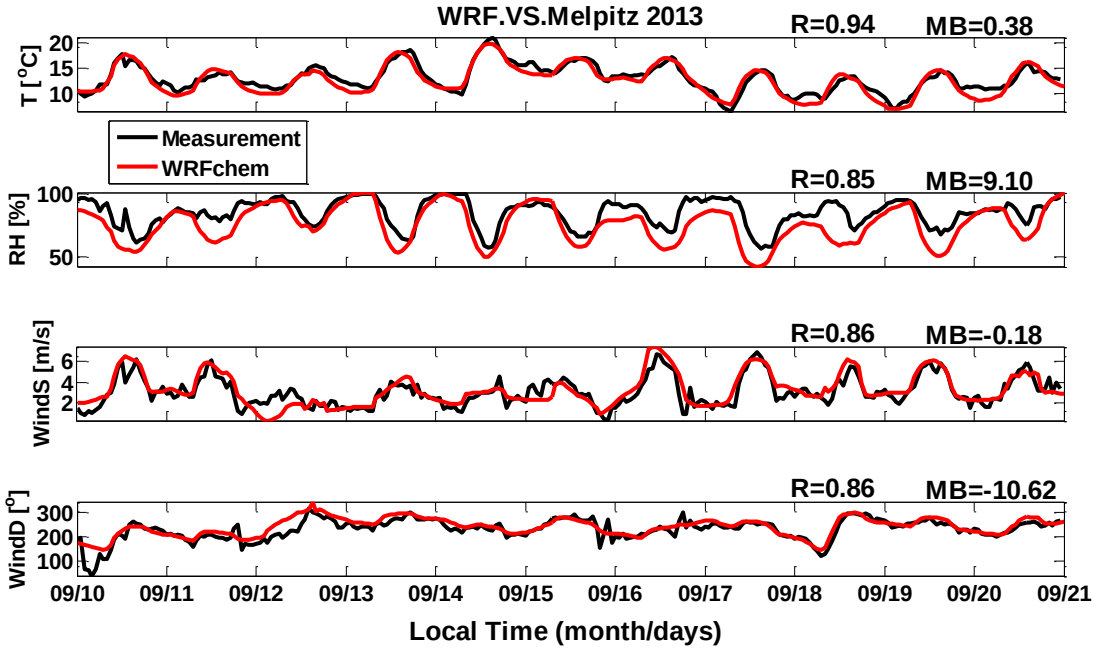


Figure S4 (newly added in the revised version). *The comparisons between the simulation results and measurements at Melpitz near-ground layer. The correlation coefficient (R) and mean bias (MB) are marked on the top of each panel. (a) Temperature; (b) relative humidity (RH); (c) wind speed; (d) wind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

The spatial patterns are also well represented by the model, as discussed in General Comments Point-2.

(2) P 5, l 42: How well match observed and simulated concentrations of the small particles?

Response:

Thanks for the comments. The sentence has been revised to discuss the results of the small particles. As shown following:

"The model significantly overestimated the concentration for the size bins 05-08 (625-10,000 nm)." ***changed to:***

"Although the simulation of PNSD/PMSD for size bins 01-04 not exactly matched with the measurements, the agreement is in the reasonable range with a factor of ~2 (Fig. 3). But the model significantly overestimated the concentration for the size bins 05-08 (625-10,000 nm) in the F-CASE."

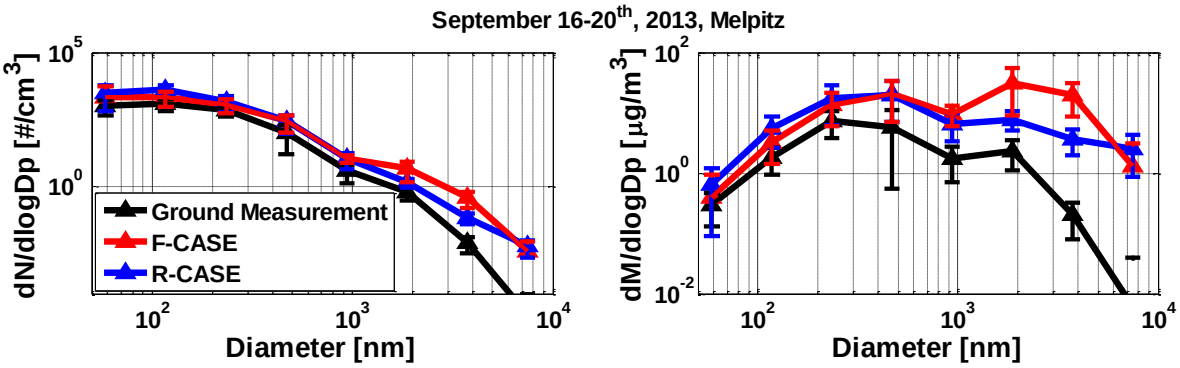


Figure 3. *Comparison of Particle Number Size Distribution (PNSD, left) and Particle Mass Size Distribution (PMSD, right) between the simulations and Melpitz measurements. The results are averaged during September 16-20, 2013; the error bars indicate the upper and lower limits.*

(3) P 6, l 1 and2: Please give some more evidence for this statement.

Response:

Thanks for the comments. The statement has been revised as suggested by the reviewer 2#. As shown below:

"Since the meteorology was well reproduced by the model, it can be assumed that the air movement was also reasonably simulated. As a consequence, there might be unrealistic high sources of coarse particles leading to the overestimation." ***changed to***

"Since the meteorology was well reproduced by the model, it can be assumed that the air movement was also reasonably simulated. Therefore, unrealistic high sources of coarse particles might be the cause for the overestimation, which would be discussed in following."

(4) P 6, l 43: How was the PBL height estimated?

Response:

Thanks for the comments. The YSU (Hong, 2006)PBL scheme is used in this WRF-Chem study. The PBL height in YSU boundary layer scheme is related to the turbulence diffusion, which keeps the basic concept of HP96 (Hong and L., 1996) but additionally includes an asymptotic

entrainment flux term at the inversion layer. And the definition of PBL height in YSU scheme is shown in following, as described by the developer:

"The PBL height is defined as the level in which minimum flux exists at the inversion level, whereas in HP96 it is defined as the level that boundary layer turbulent mixing diminishes."

More detailed information about this YSU boundary layer scheme is given in (Hong, 2006)

(5) P 7, l 9: According to Fig. 6b, the sea salt layer does not yet touch the surface. What is the contribution of turbulent mixing after sunrise?

Response:

It is a very good question. One figure (Fig. 6c) and the corresponding discussion have been added, in order to answer this question. As shown following:

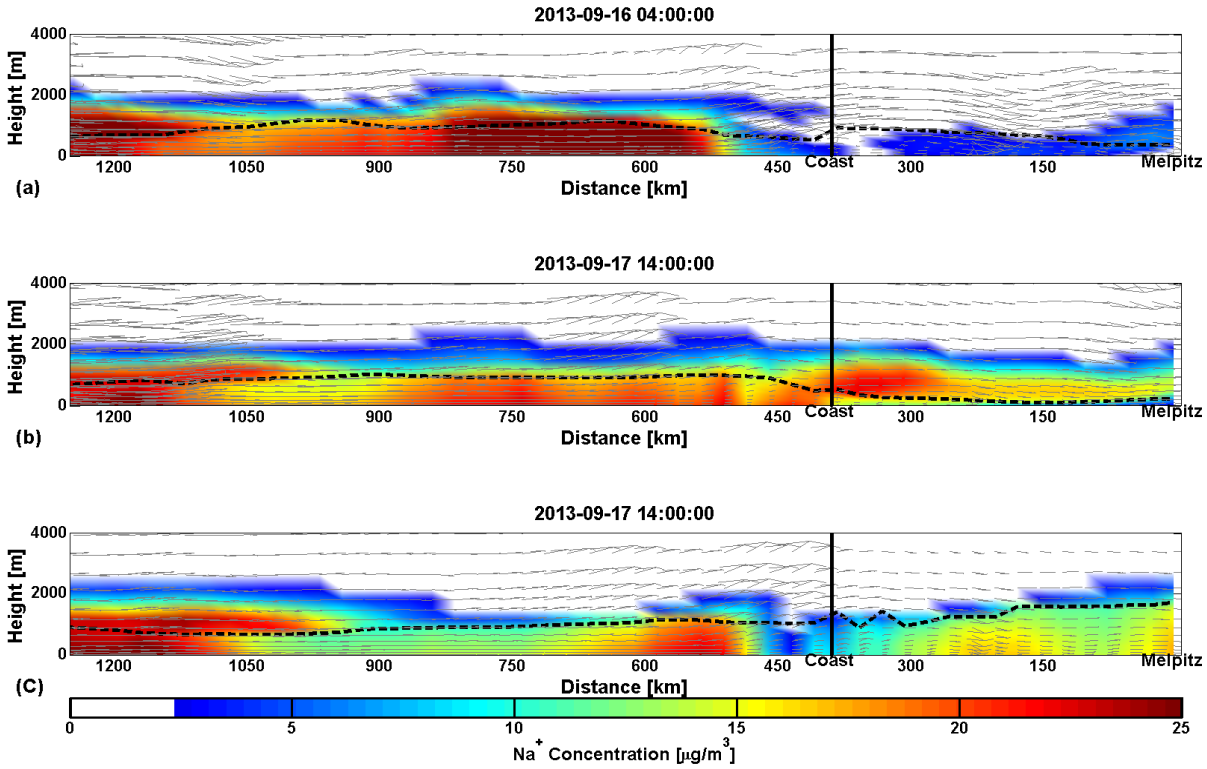


Figure 6. *WRF-Chem result of the sea salt (Na^+) concentration on the vertical cross section, which is shown by the black dash line in Figure 1. The locations of Melpitz and coast (black line) are marked. The grey arrows indicate the wind field, and the black dash line indicates*

the planetary boundary layer (PBL) height. (a) 2013-09-16, 04:00 LT; (b) 2013-09-17, 02:00 LT; (c) 2013-09-17, 14:00 LT.

"Then the downward draft, resulted from high-pressure ridge, brought the lofted SSA back into the surface layer (Fig. 6b)." ***changed to***

"Then the downward draft resulted from high-pressure ridge and the turbulent mixing after sunrise (Fig. 6b and 6c), brought the lofted SSA back into the surface layer. The Na^+ mass concentration at Melpitz surface increased from ~7 $\mu g/m^3$ (Fig. 6b) to ~15 $\mu g/m^3$ (Fig. 6c). About 35% of the lofted SSA contributed to the increase of the Na^+ surface concentration. This result is agreement with the previous study (Chen et al., 2009), which reported ~30% of elevated pollutants contributed to the increase of surface pollutants concentration in Beijing, due to the turbulent mixing after sunrise."

(6) P 7, l 25 and 26: There could be also some other reasons, wrong turbulent exchange, wrong water uptake (also due to wrong relative humidity), . . .

Response:

Thanks for the comments. Yes, the reviewer is correct. The corresponding sentence has been removed.

(7) P 7, L 29: Why can this be expected?

Response:

Thanks for the comments. It is because: assuming that the chemical mechanism is correctly described in the model, the same concentration of gaseous precursors (NOx and ammonia) should produce the same concentration of nitrate in the model and observation. In order to describe this point more clearly, the paragraph has been revised as suggested by reviewer 2#. As shown below:

"The location of the data dots (Fig. 7a) may be shifted due to the uncertainty of precursors emissions, but the nitrate mass concentration is always expected to be consistent with the observed concentration in Fig. 7b. The difference between Fig. 7a and Fig. 7b indicates that

in addition to an overestimation caused by overestimated 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 though where had the same mass concentrations of precursors." ***changed to***

"The location of the data dots (Fig. 7a) may be shifted due to the uncertainty of precursors emissions, but the nitrate mass concentration is always expected to be consistent with the observed concentration in Fig. 7b when they have the same mass concentration of precursors. However, even under the same mass concentrations of precursors, the simulated nitrate mass concentrations (Fig. 7a) were still significantly higher than the observed ones (Fig. 7b). This indicated that in addition to an overestimation caused by overestimated NH3 (see also Table 2), improper chemical pathway in the model also contributed to the nitrate overestimation."

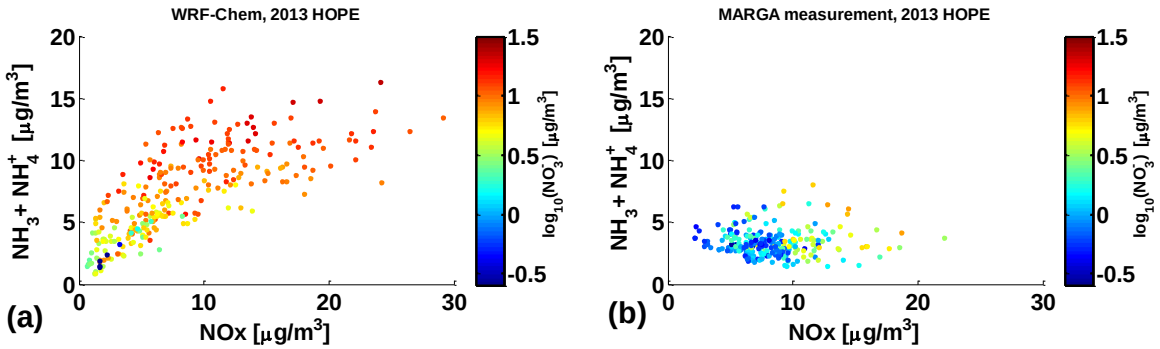


Figure 7. *Relationship between nitrate, total ammonia and NOx during September 10-20, 2013 at Melpitz. The color indicates the nitrate mass concentration in logarithmic scale. (a) WRF-Chem model results; (b) MARGA measurement results.*

(8) P 7, l 1 – 10: Please change the order of the figure, Figure 9 should be discussed here.

Response:

Thanks for the comments. The Figure 9 has been revised to include the impact of sea salt on nitrate particle mass size distribution (PMSD), resulting from the heterogeneous reaction on sea salt surface. The revised Figure 9 (please find it in General Comments Point-1) would help this manuscript make a more clear conclusion and connect the scientific points together. Therefore, we move the revised Figure 9 to the end of section 3.4. As shown below:

"In general and as illustrated in Fig. 9, the overestimation of SSA emission scheme has a significant influence on the particulate nitrate simulation in both the coarse mode (directly) and the fine mode (indirectly)."

(9) P 8, l 8: Is this really a probability distribution or a frequency distribution?

Response:

Thanks for the comment. It was really a probability density function. But now, we have modified it as suggested by the reviewer 1#. The "probability density function" has been replaced by "frequency distribution", the patterns are the same between them two. As shown below:

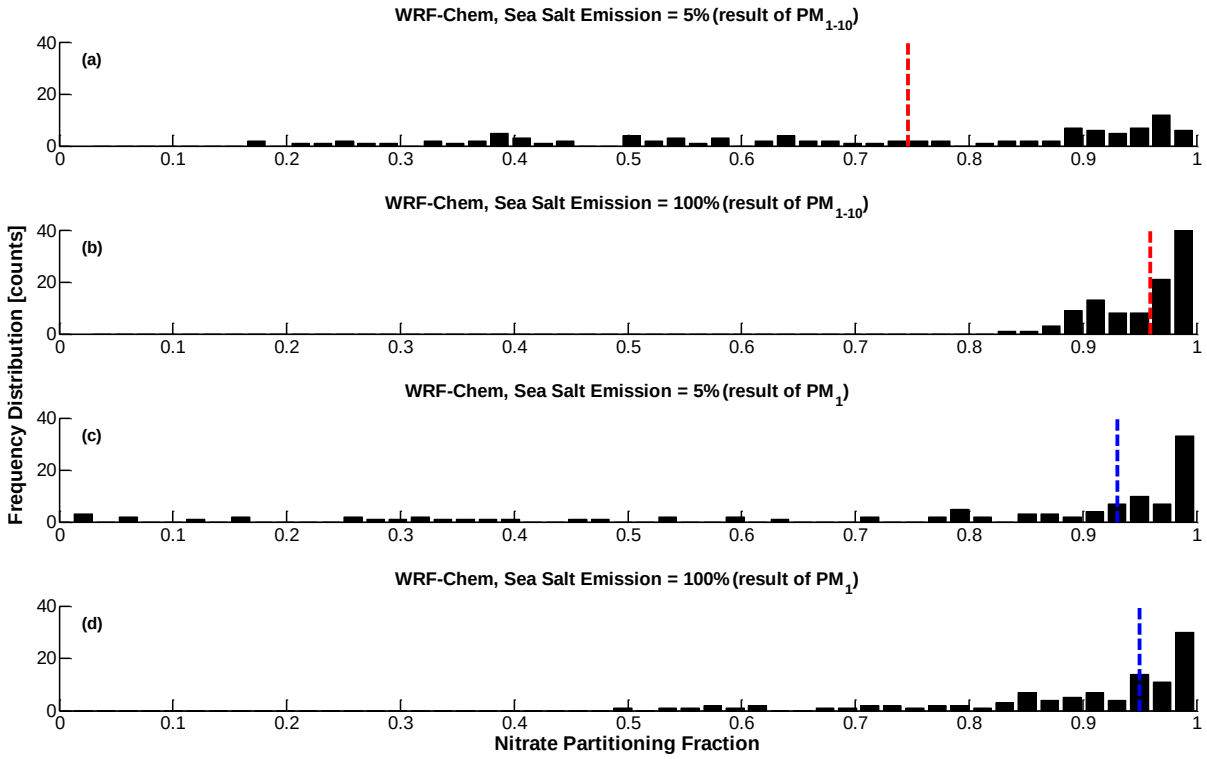


Figure 8 (revised). *WRF-Chem results of the frequency distribution of PF_nitrate at Melpitz. The result was analyzed during the marine period ($[Na^+]$ > 1.8 μg/m^3 in the F-CASE). The dash lines (coarse mode: red; fine mode: blue) indicate the median value (with 50% probability in both sides). (a) PM_{1-10} result of 5% SSA emission (R-CASE); (b) PM_{1-10} result of the F-CASE; (c) PM_1 result of the R-CASE; (d) PM_1 result of the F-CASE.*

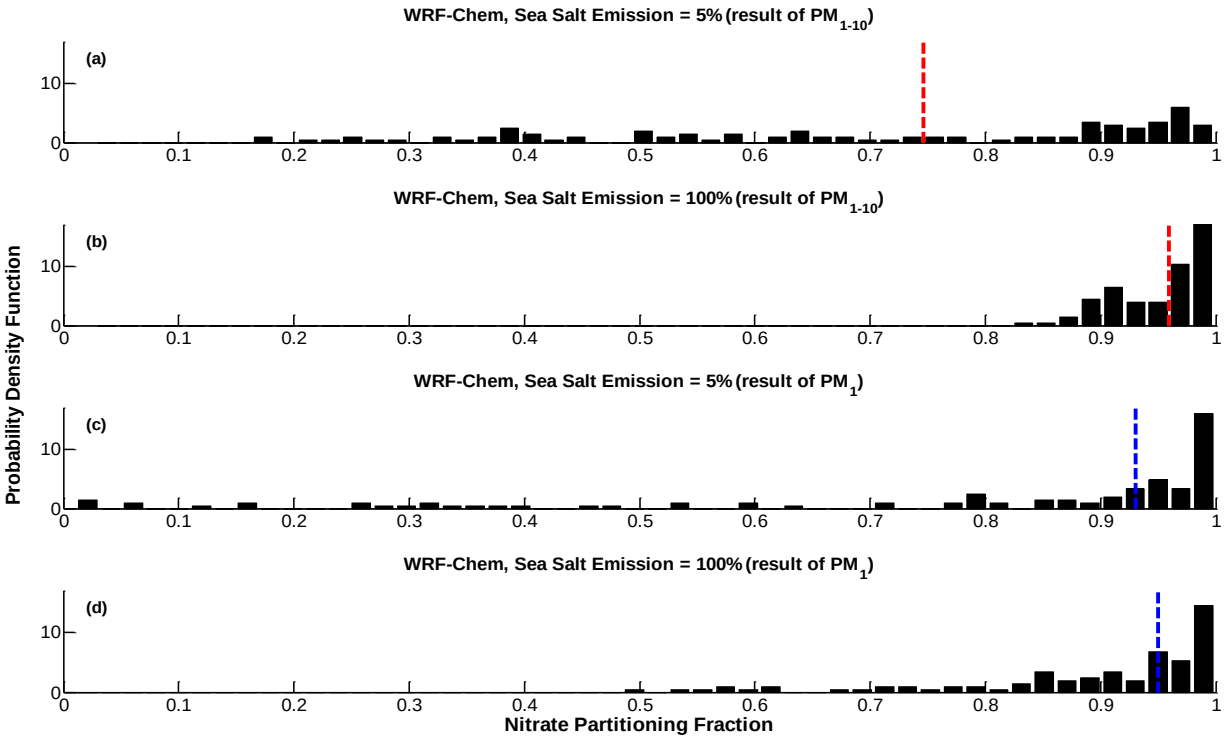


Figure 8 (original). *WRF-Chem results of the probability density function of nitrate partitioning fraction (PF_nitrate) at Melpitz in the marine period during September 10-20, 2013. The marine period is defined as the Na^+ mass concentration higher than 1.8 $\mu g/m^3$ in the F-CASE. The blue dash lines indicate the median value (with 50% probability in both sides). (a) PM_{1-10} result of 5% SSA emission (R-CASE); (b) PM_{1-10} result of the F-CASE; (c) PM_1 result of the R-CASE; (d) PM_1 result of the F-CASE.*

(10) Figure 1 and Figure 6: Please consider using a different color scheme. In particular, the dark blue color for the low values is quite unfavorable and the blue arrows (and the map in Fig. 1) can hardly be recognized.

Response:

Thanks for the comments. The color schemes in Figure 1 and Figure 6 have been changed. Please find the revised Figure 6 in the Detailed Comments Point-5. And the revised Figure 1 is shown below:

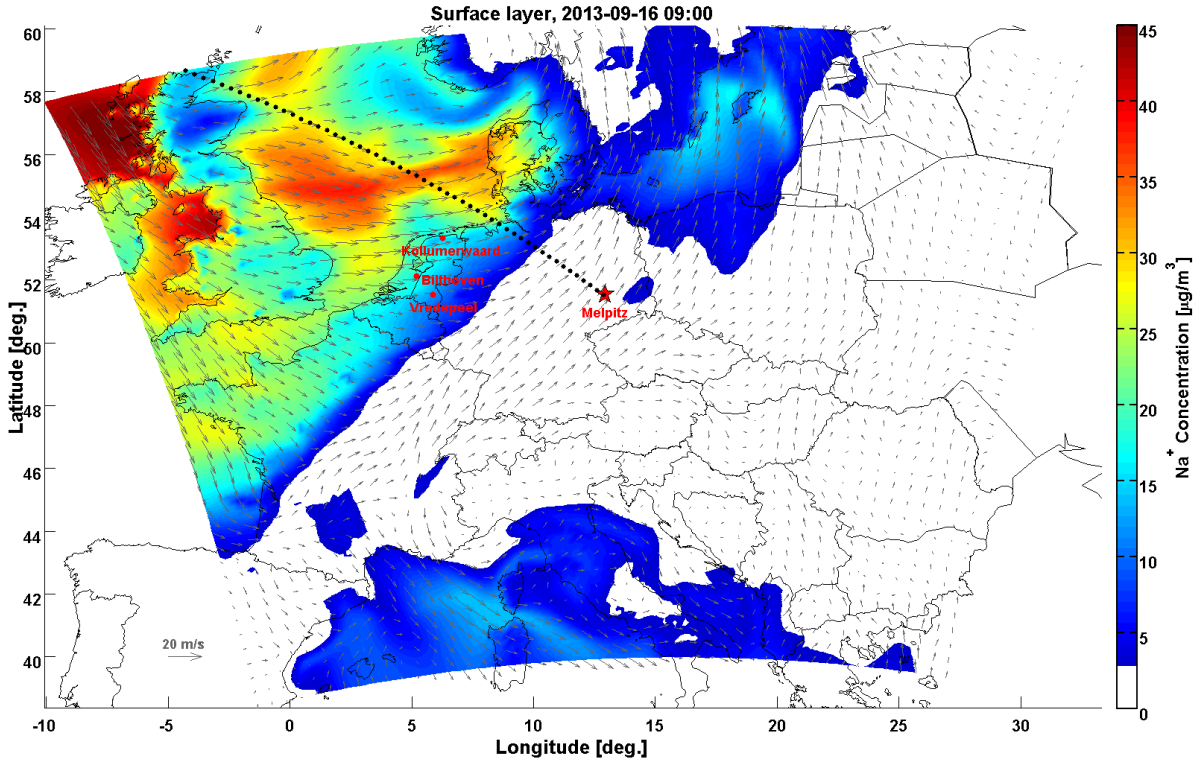


Figure 1. *The horizontal distribution of surface Na^+ mass concentration in domain 02 (intermediate domain) at 2013-09-16, 09:00 LT. The grey arrows indicate the wind field. The locations of 4 EMEP stations (Melpitz, Bilthoven, Kollumerwaard and Vredepeel) are marked. The vertical cross section of dash black line is shown in Figure 6.*

(11) Figure 3: Please show also the R-case.

Response:

Thanks for the comment. The R-CASE result has been added as suggested. Please find the revised Figure 3 in the Detailed Comments Point-2. Comparison between the F-CASE and the R-CASE in Figure 3 also partly supported the scientific point, that the overestimation of SSA inhibited the fine mode nitrate particle formation. This information has been added in section 3.4, as shown below:

"In order to see the influence of SSA on nitrate PMSD in a more clear 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).”

(12) Caption of Fig. 4: Please mention which case is shown.

Response:

Thanks for the comment. The caption of Figure 4 has been revised as shown below:

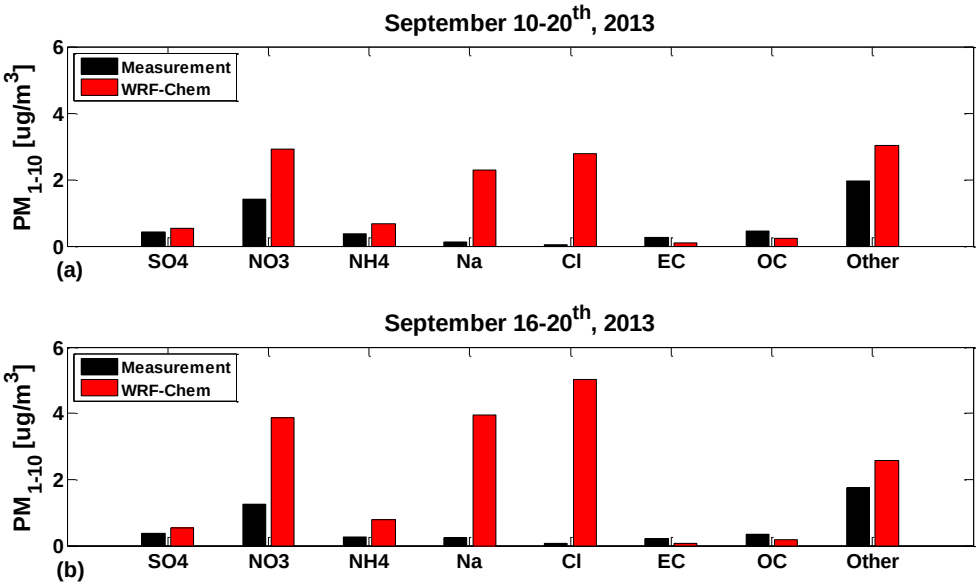


Figure 4. *Comparison of coarse mode aerosol ($PM_{1\text{-}10}$) chemistry compounds between the* ***F-CASE*** *results and Melpitz measurements. (a) averaged during the HOPE-Campaign period of September 10-20, 2013; (b) averaged during the marine air mass period of September 16-20, 2013.*

(13) Caption of Fig. 5: Please mention particle size. Please mention the different scale for observations and model results.

Response:

Thanks for the comment. The caption of Figure 5 has been revised as shown below:

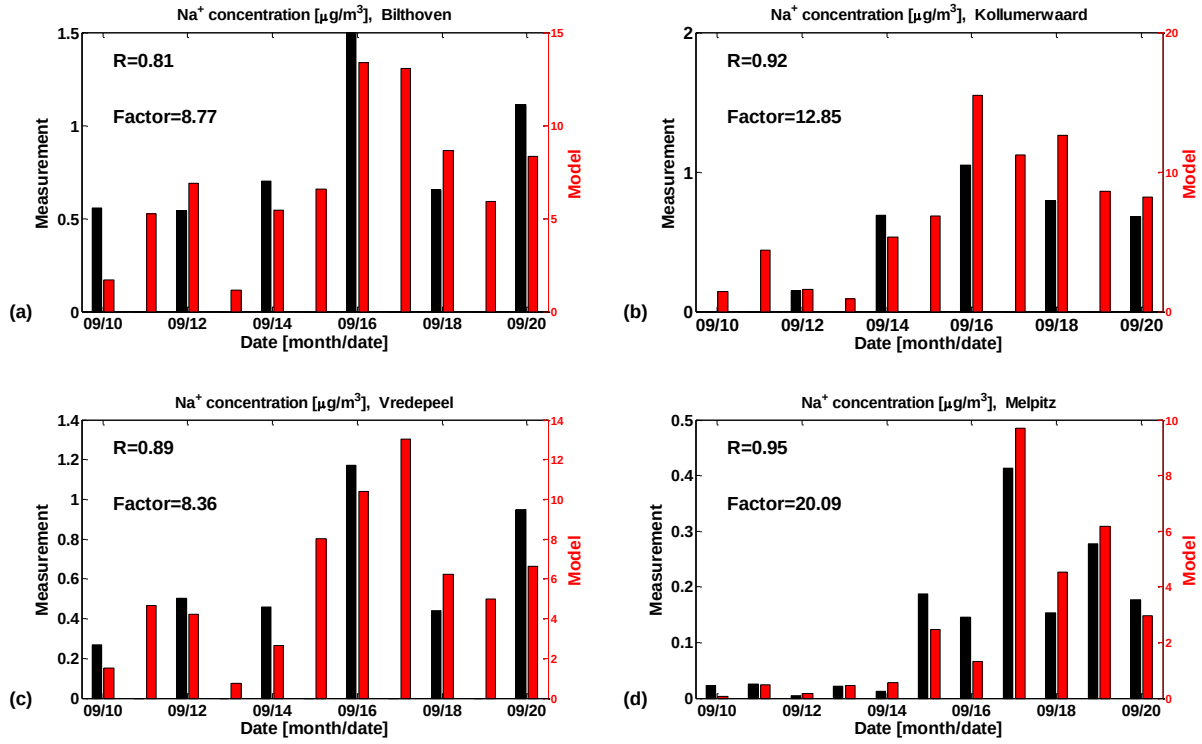


*Figure 5. Comparison of Na^+ mass concentration **in PM_{10}** between the filter sampler measurements **(left y-axis)** in 4 EMEP stations and **the F-CASE** results **(right y-axis).** (a) Bilthoven; (b) Kollumerwaard; (c) Vredepeel; (d) Melpitz. The locations of stations are shown in Figure 1.*

(14) Figure 6 and (current) Figure 9: These figures should be oriented from West (left) to East (right). No need for the star, as Melpitz is located at the Eastern end of the figures.

Response:

Thanks for the comment. The Figure 6 and Figure 9 have been revised as suggested by reviewer and the reviewer 1#. Please find the revised Figure 6 in the Detailed Comments Point-5, and the revised Figure 9 in the General Comments Point-1.

Minor issues:

(1) P 2, l7: Partitioning is no 'formation'.

Response:

Thanks for the comment. The sentence has been revised as shown below:

"SSA could participate in heterogeneous reactions by interacting with trace gases, leading to the formation of secondary aerosols (Seinfeld, 2006)," ***changed to***

"SSA could participate in heterogeneous reactions by interacting with trace gases, leading to the formation of secondary aerosol particles on SSA surface (Seinfeld, 2006),"

(2) P 2, l 13: '. . . sodium nitrate is largely contributed to nitrates': please reword.

Response:

Thanks for the comment. The sentence has been revised as suggested by the reviewers 1&2#. As shown below:

"sodium nitrate is largely contributed to nitrates in northern and southern Europe" ***changed to:***

"sodium nitrate largely contributes to nitrates in northern and southern Europe"

(3) P 2, l 21, 22: 'opportunity' and 'make their influence more extensive': please reword

Response:

Thanks for the comment. The sentence has been revised as suggested by the reviewer 2#, as shown below:

"These mechanisms provide an opportunity for SSA to be transported inland, and thereby make their influence more extensive, from coastal to regional or even global." ***changed to***

"These mechanisms provide make the long-range transportation of SSA easier, and thereby could expand their influence range from coast to a broader region."

P 2, l 33: Southern ???

Response:

Thanks for the comment. The "Southern" has been revised to "South Pacific"

P 2, l 43: influence on what?

Response:

Thanks for the comment. The "influence of SSA" has been revised to "influence of SSA on the size resolved nitrate particle formation".

P 3, l 27: Please mention first that a resistance approach is applied.

Response:

Thanks for the comment. The sentence has been revised as shown below:

"The dry deposition of particles is calculated on the basis of the sublayer resistance, aerodynamic resistance and surface resistance (Grell et al., 2005)." ***changed to***

"The dry deposition of particles is calculated by ***a resistance approach****, including sublayer resistance, aerodynamic resistance and surface resistance (Grell et al., 2005)."*

P 6, l 10: Please reword: an event cannot be emitted.

Response:

Thanks for the comment. The "abrupt SSA event was found to be emitted over the North Sea" has been revised to "abrupt SSA emission event happened over the North Sea".

P 7, l 33: A word seems to be missing here.

Response:

Thanks for the comment. The sentence has been revised as suggested by the reviewer 2#. As shown below:

"The difference between Fig. 7a and Fig. 7b indicates that in addition to an overestimation caused by overestimated 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 though where had the same mass concentrations of precursors." ***changed to***

"However, even under the same mass concentrations of precursors, the simulated nitrate mass concentrations (Fig. 7a) were still significantly higher than the observed ones (Fig. 7b). This indicated that in addition to an overestimation caused by overestimated NH_3 (see also Table 2), improper chemical pathway in the model also contributed to the nitrate overestimation."

P 9, l 38: the last sentence is incomprehensible.

Response:

Thanks for the comment. The sentence has been removed as suggested by the reviewer 1#.

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