Response to comments of referee #1: Dr. Daniel Neumann

General comments

The submitted manuscript describes the impact of sea salt aerosol on atmospheric nitrate concentrations. The presence of sea salt aerosol leads to a mass-enhancement of nitrate in the particle phase but also to a re-distribution of nitrate from fine to coarse particles modes. However, the two processes increase and decrease the absolute value of nitrate direct radiative forcing, respectively, and, hence, are in competition to each other. In theory, the considered impact of particulate sea salt on the size distribution of particulate nitrate and the resulting change of the DRF is well understood. Practically, however, we do not have exact numbers on how strong the impact is. The authors evaluate this competition for certain environmental conditions and spatial scales. The manuscript is well written and structured. The scientific aim is clearly formulated. The main manuscript provides the relevant information as brief as possibles, whereas an extended presentation of the model description and results are provided in the supplement. I appreciate the quite detailed supplementary text and the additional figures. I have several comments on the manuscript. However, these are no critical aspects. In summary, I recommend the manuscript for acceptance after minor revision.

Many thanks to Dr. Neumann for the careful reading and the helpful comments and suggestions. We have improved the manuscript accordingly. Please find a point-by-point response below. Please refer the order of figures to the revised version.

Major comments:

1) What are the statistics of the sea-salt-over-continent-transpor events in Europe? Do they take place sufficiently often and/or do they persist over a sufficient long time period in order to have relevant impact on the annual mean $DRF_{nitrate}$?

Marine air masses influence Europe quite frequently. In a previous long-term observation-based study (Birmili et al., 2001), they classified the air masses influencing Central Europe as 'Maritime Character', 'Continental Character' and 'Mixed-Type Character'. They performed a statistical study with observations of more than one year and reported that the frequency of

'Maritime Character' and 'Mixed-Type Character' air masses are 46.6% and 33.3%, respectively (see the Table 1b of Birmili et al. 2001). Therefore, the marine air masses influence Europe with a total frequency of about 90%, and are expected to have impact on the annual mean $DRF_{nitrate}$ by such a high frequency. We have modified the summarization of the frequency in section 3.1 to make it clearer, as shown below.

"Marine air masses frequently (~90%) approach Central Europe (Birmili et al., 2001). The interaction between anthropogenic pollutants and sea-salt aerosol commonly happens in the atmosphere."

Changed to:

"Marine air masses frequently, up to 90% of days in a year, influence Central Europe (Birmili et al., 2001). The interaction between anthropogenic pollutants and sea-salt aerosol commonly happens in the atmosphere."

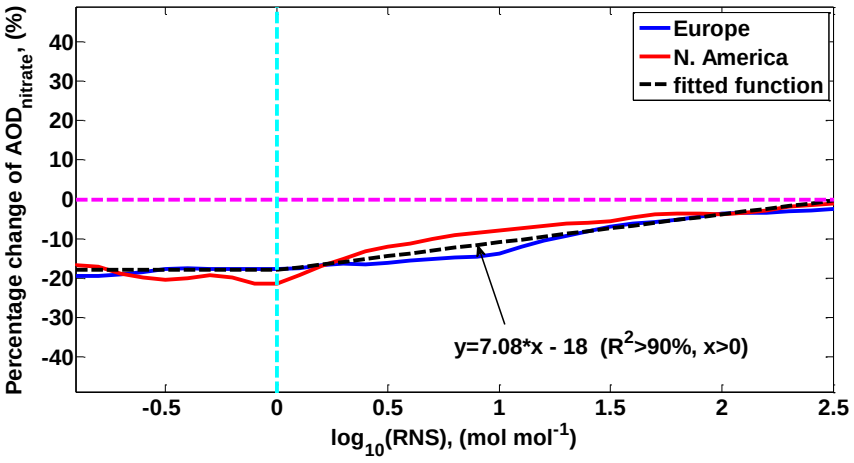
2) For the European modeling period a time frame containing a strong sea-salt-transport event was choosen. For the US example, an arbitrary time period was choosen. How far are both modeling cases – Europe and US – comparable?

There are similarities and differences between the European case and the US case.

The difference. For Europe case, a sea-salt event was chosen, we see a strong 're-distribution effect'. This leads to a reduction of 10-20% in $AOD_{nitrate}$ over European polluted regions, such as northern polish region, and a clear reduction of $AOD_{nitrate}$ over most of European continent (see Fig. 5a). For US case, we see outflows bring continental pollutants to marine and a clear 're-distribution effect' leading to reduction of $AOD_{nitrate}$ over oceanic regions, such as the Gulf of Mexico; but, there is much weaker 're-distribution effect' over the N. American continent (see Fig. 8a). This difference between the European and US cases indicates that 're-distribution effect' happens wherever natural sea-salt and anthropogenic nitrate mixed, no matter over continental or oceanic regions. Lots of big cities are along coast globally with high NOx (precursor of nitrate) emissions, offshore and onshore wind patterns can both induce the 're-distribution effect' and reduce $AOD_{nitrate}$.

The similarity. Although we see different spatial patterns of the 're-distribution effect' in the European and US cases, the relationships between $AOD_{nitrate}$ reduction and RNS (molar ratio

between nitrate and sodium) are quite consistent in these two cases. As suggested by the Reviewer#2, we show this relationship as a first-order approximation, given in the newly added Fig. 9b. Later on, we adopt this generalized relationship combined with EMAC global model results to estimate the potential changes of $AOD_{nitrate}$ associated with 're-distribution effect' on a global scale.



Newly added Fig. 9b. *The median possibility of the percentage change of $AOD_{nitrate}$ as a function of RNS.*

3) On p.4 l.116 the authors state that they found in previous studies that – using the Gong emission parameterization – modeled sea-salt concentrations overestimated measurements by a factor of 10. However, the sea salt emissions estimated by the Gong parameterization are not necessarily 10x as high as the real world sea salt emissions are. Processes that lead to lower atmospheric sea salt concentrations might be underestimated in the used model – e.g. underestimated deposition. Or, the particle size distribution of the Gong sea salt emissions might not be appropriate for the model setup. Therefore, please add ". . . using WRF-Chem . . . " or something similar to the sentence – e.g. in the end of the sentence.

We have modified the context as suggested.

4) In the first paragraph on page S9 in the supplement, the authors discuss that nitrate concentrations are overestimated by several models in Europe. Vivanco et al. (2017, DOI: https://doi.org/10.1016/j.atmosenv.2016.11.042) evaluated several air quality models and found that most considered models underestimate the nitrogen deposition in Europe (The

EMEP model performed best). WRF-Chem was not partof the latter evaluation. However, too high atmospheric nitrate concentrations may point to issues in the deposition parameterizations. This wouold also explain why the Gong parameterization apparently lead to considerably overestimated atmospheric sea salt concentrations. Could the authors comment on that?

This is a good point, thanks for the suggestion. We have added a comment in the last paragraph of section 3.4, as shown below.

"The uncertainty in deposition parameterizations could be one reason of this overestimation, as suggested by the overestimation of sea-salt particles as well (Chen et al., 2016a) and the underestimation of nitrogen deposition over Europe in many models (Vivanco et al., 2017)."

5) Evaluating the mass concentration of sea salt aerosol is reasonable to evaluate the coarse sea salt emissions. The contribution of fine sea salt to the total sea salt mass concentration is very low. Hence, the strong overestimation of sea salt mass concentrations documented in previos studies does not necessarily indicate that the fine particulate sea salt was overestimated as well. The black bars in Fig. S3 rather indicates that fine sea salt might be underestimated in some episodes. Please briefly describe this uncertainty in the Materials & Methods or in the Discussion section.

Good point. We have added discussion of this uncertainty in the Materials & Methods, section 2.2, as shown below.

"The sea-salt emissions computed with the modified Gong scheme (Gong, 2003) were reduced to 10% in the 'Case_SeasaltOn', because a previous study (Chen et al., 2016) has shown that the original Gong scheme overestimates the sea-salt mass concentrations by a factor of ~10 over the coastal regions of Europe using WRF-Chem model. We note that although the mass of coarse sea-salt particles is certainly overestimated, it might not necessarily indicate overestimation in fine sea-salt particles related to their minor contribution to the total mass."

6) p.6 l.165-168 "In addition, a one-year simulation with global model (EMAC) was carried out for analysis of the potential impact of 're-distribution effect' on a global scale, although the fully dynamic mass transfer between particle sizes is not considered in EMAC (four size modes

rather than eight size bins as applied in the WRF-Chem model).” Why is EMAC neverthelss applicable for the used purpose?

Here, we did not directly use EMAC model to estimate the impact of ‘re-distribution effect’. We generalized a relationship between the impact of ‘re-distribution effect’ and RNS (molar ratio between nitrate and sodium) from the European and US cases, and then used this relationship combined with the RNS values from EMAC model to investigate the potential impact of ’re-distribution effect’ on a global scale. Please also see the ‘The similarity’ part of the response in point-2. We have modified the context to make it clearer, as shown below.

“In addition, a one-year simulation with global model (EMAC) was carried out for analysis of the potential impact of ‘re-distribution effect’ on a global scale. Although the fully dynamic mass transfer between particle sizes is not considered in EMAC (four size modes rather than eight size bins as applied in the WRF-Chem model), we adopt a parameterization derived from WRF-Chem simulations to estimate the potential impact (details given in section 3.5).”

7) “Sea-salt is emitted into the marine planetary boundary layer (PBL) as coarse particles [. . .]” (p.7 l.183). Aren’t it coarse and fine particles? Depending on the considered moment of the particle size distribution the fine or the coarse mode dominates: the number distribution is dominated by the fine mode; the volume/mass distribution is dominated by the coarse mode. Please clarify this in the text.

Yes, the reviewer is right that it has to be precise. We have clarified this, as shown below.

“Sea-salt is emitted into the marine planetary boundary layer (PBL) with mass concentration dominated by coarse particles…”

8) The authors mention the importance of vertical transport of sea salt aerosol into the continental free troposphere followed by the horizontal transport of it over land and refer to their previous studies (p.7 l.186-190). This transport mechanism seems to be very important. However, vertical transport/convection is not necessarily well represented by all meteorological models – depending on grid resolution and parameterization. This study’s

WRF-Chem model simulations probably reproduce this vertical transport of sea salt? Do the other used models reproduce it as well?

The vertical transport of sea-salt particles to free troposphere can increase their lifetime and facilitate their inland transport, therefore, it could be important for the 're-distribution effect' over the deeper inland regions. This vertical transport and the associated land inward transport are reproduced well in our high-resolution simulation using WRF-Chem, as shown in our previous study (Chen et al., 2016). However, as the reviewer concerned, this vertical transport may not be well reproduced in other models with lower resolutions. But, this does not necessarily mean that sea-salt cannot transport further inland in other models. The long-range transport of sea-salt to inland regions also depends on the deposition scheme (which might be underestimated as pointed out by the reviewer in the point-4), the vertical mixing rate, the parameterizations for marine boundary layer and continental boundary layer and etc. These can be different model-by-model and case-by-case, and the discussion of these differences go beyond the scope of this study.

But, I think the vertical transport might not be such important for the coastal regions, where lots of big cities with high NOx emissions are located, especially for China and N. America with many large and the most developed cities along the coast. This is because no matter offshore or onshore wind patterns both mix sea-salt with anthropogenic nitrate and induce the 're-distribution effect', as discussed in the point-2.

9) In chapter 3.3 (p.10 l.281-290), the authors mention the aim to 'generalize' the results. To 'generalize' something has a quite broad meaning and can be interpreted differently. The benefit of calculating the RNS and of including Fig. 5 did not become clear for me in chapter 3.3 – but, later in chapter 3.4 it became obvious. Please clarify the motivation in chapter 3.3.

We have modified the corresponding sentence and clarify the motivation in section 3.3, as shown below.

"To compare the relative importance of the sea-salt-induced 're-distribution effect' and 'mass-enhancement effect' on anthropogenic nitrate cooling…"

10) Classification of RNS: Currently, we have the situations “RNS < 1”, “1 < RNS < 30” and “30 < RNS”. I know that these are rough classifications. Nevertheless, please also cover “RNS == 1” and “RNS == 30” to be mathematically correct.

Thanks for the carefulness. We have corrected it as suggested.

Comments on figures:

1) Figure 1: I like the intention of Fig. 1. All relevant aspects are shown. However, on the first view, the reader might not recognize the particle size distribution plot as such. At least I did not recognize it first. I am not sure how to improve the Figure. Possibly, a y-axis on the left of the plot might help. Their are no issues when the figure is printed in grey-scales (see my comments to the other figures below).

We have added a y-axis in Fig. 1. It does make the particle size distribution plot clearer. Thanks.

2) Figure 2, 6, 8 and 9: Please consider using a color scale, which is recognizable in black-and-white and readable by color blind people (not “jet” or “rainbow”). Examples for such a color scale are “viridis” and “magma”.

We have changed the color scales of Fig. 3, Fig. 7, Fig. 9 and Fig. 10 (Fig. 2, Fig. 6, Fig. 8 and Fig. 9 in the original version) to make them recognizable in grey-scales.

3) Figure 4 and 7: Printed in grey scales it is hard to distinguish whether values are negative or positive. However, alternative color scales would make the full-color plots difficult to interprete. Therefore, it might be reasonable to keep the blue-white-red colorscale.

We did not find a proper color-scales fit in the grey-scales perfectly, and also clearly show the trends. Agree with the reviewer, it is better to keep it as it is.

4) Figures 3: Please add information to the plot that nitrate is plotted. It is written in the caption but it would be nice to also have this information in the plots. The colors in this figure are fine.

Done. 'Nitrate' has been added in the y-axis.

5) Figures 5: The colors in this figure are fine. Figure S6: Maybe exchange panels (a) and (b). For me it would have made the figure easier to understand. I would consider adding Figure S6 to the main manuscript.

Thanks. As suggested, we have moved the Fig. S6a (original) to Fig. 7c (revised), and Fig. S6b (original) to Fig. 7b (revised).

Minor Comments:

p.4 l.117: consider to replace "for both with and without sea-salt presence respectively" by "for both sea salt emission cases"

p.5 l.123: remove "and" in ". . . assumption and taking . . . "

p.5 l.136: "and part of North Africa" → "and the northern part of North Africa"

p.5 l.142 "1/8_ × 1/16_" → space between "×" and "1/16"

p.5 l.147 "In the 'Case SeasaltOn' (with sea-salt emission) of European simulation" → "In the European 'Case SeasaltOn' simulation (with sea-salt emission)", suggestion

p.5 l.148 "over coastal region" → "over coastal regions", added plural-s

p.6 l.149/150 "with a factor (and correlation coefficient) of 0.85 (0.67), 1.16 (0.80) and 0.83 (0.87) respectively for Bilthoven, Kollumerwaard and Vredepeel (Fig. S2)" → "with a factor (and correlation coefficient) of 0.85 (0.67), 1.16 (0.80) and 0.83 (0.87) for Bilthoven, Kollumerwaard and Vredepeel, respectively (Fig. S2)", moved 'respectively'

p.6 l.154 “over coastal, German low lands (Melpitz) and northern Poland regions” → “over coastal, German low land (Melpitz) and northern Polish regions”, remove ’s’ from lands because we have “coastal regions”, “German low land regions” and northern Polish regions”

p.6 l.162 “concentration of nitrate was” → “concentrations of nitrate were”

p.6 l.164 “covers US, the Gulf of Mexico and part of Pacific and Atlantic oceans” → “covers the US, the Gulf of Mexico and parts of Pacific and Atlantic oceans”, ’the’ in front of ’US’ and ’s’ added to ’part’

p.6 l.171 “1.1 by 1.1 degrees” → “$1.1^{o} \times 1.1\ ^{o}$”, formatting consistent with p.5 l.142

p.6 l.161 “boarder” → “broader”

p.6 l.177 “approach Central Europe, and the interaction” → “approach Central Europe.

The interaction”

p.7 l.179 “a typical sea-salt transport event”; maybe ’sea salt transport event’ in italics

p.7 l.182 “originated” → “originating”

p.7 l.187 “long-range transport, see the figure 11 of Chen et al. (2016a)” → “long-range transport (see Fig. 11 of Chen et al. (2016a))”

p.8 l.212: “the northern Poland region” → “a region in northern Poland”

p.8 l.220: “,thus ” → “, thus, ”

p.8 l.227: “Over the northern Poland region” → “Over the region in northern Poland”

p.8 l.231-234: “being slightly . . . in the offline calculation” – Please consider writing this sentence outside of the brackets. It improves the readability.

p.9 l.241-242: “with increasing AOD with [NO3-]” → “of increasing AOD with increasing [NO3-]”; I am not sure about it but for me it sounds better.

p.9 l.242: “would result by neglecting the ’re-distribution effect”’ → “would result when t he ’re-distribution effect’ was neglected”

p.9 l.242-244: “For example, instead of a decrease by 29

p.9 l.245: remove “also”

p.9 l.245: “. . . increases and there is . . . ” → “. . . increases. There is . . . ”

p.9 l.248: “and thus shorter” → “and, thus, shorter”

p.9 l.251: “and hence further moderate” → “further moderating”

p.9 l.254: “, therefore fasten” → “fastening”

p.9 l.255: “oxygenated nitrogen” → maybe “oxidized nitrogen”?

p.9 l.257: “which result” → “which resulted”

p.9 l.258-263: This has been mentioned previously. Although, this is important motivation for performing another simulation without deposition, I would like to suggest shortening these sentences considerably.

p.10 l.265-266: “as the simulations with aerosol dry deposition” → “as in the simulations with aerosol dry deposition turned on”; corresponds better with l.263-264 stating “. . . with aerosol dry deposition turned off”

p.10 l.289: “due to” → “because”; ’due to’ does not allow a verb (’are’)

p.11 l.319-320: “Myhre et al. (2006), where a similar . . . aerosol was evaluated but the simplification . . . ” → “Myhre et al. (2006). They evaluated a similar . . . aerosol. But, their simplification . . . ”; split into two or three individual sentences;

p.12 l.332: “and thus reduces” → “and, thus, reduces”

p.12 l.337: “presented” → “present”

p.12 l.342-343: “that sea-salt transport (May et al., 2018) and impact AODnitrate further inland over North America.” → “that sea-salt is transported further inland over North America und impacts AODnitrate there (May et al., 2018).”

p.13 l.352-356: Please refer to Fig. 9 (maybe I overlooked the reference).

p.13 l.360: “and hence lower” → “and, hence, lower”; I would consider splitting the sentence into two sentences at this ’and’.

p.13 l.369: “if only consider” → “if we only consider” or passive form

p.13 l.374: “. . . when RNS is lower than 1” → Formulation is ambiguous in this context: the dominance could take place when RNS < 1 OR the mass-enhancement effect could be associated with RNS < 1. It know (and probably most readers) know that the latter is meant. However, I would suggest to reformulate this part. The same for the next sentence (“. . . when 1 < RNS < 30“).

Many thanks for the careful reading, helping correct the typos and improving the language. We have improved the manuscript as suggested. Please find details in the change-tracked revised manuscript.

References:

Birmili, W., Wiedensohler, A., Heintzenberg, J., and Lehmann, K.: Atmospheric particle number size distribution in central Europe: Statistical relations to air masses and meteorology, Journal of Geophysical Research, 106, 32005-32018, DOI: 10.1029/2000JD000220, 2001.

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Gong, S. L.: A parameterization of sea-salt aerosol source function for sub- and super-micron particles, Global Biogeochemical Cycles, 17, 10.1029/2003GB002079, 2003.

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