Atmospheric
Chemistry
and Physics

Discussions

Open Access

Interactive comment on "Natural sea-salt emissions moderate the climate forcing of anthropogenic nitrate" *by* Ying Chen et al.

Daniel Neumann (Referee)

daniel.neumann@io-warnemuende.de

Received and published: 9 September 2019

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.

1   Major comments

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 $DRF_{nitrate}$?

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?

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.

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?

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.

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?

"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.

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?

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.

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.

2 Comments on figures

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).

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".

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 interpret. Therefore, it might be reasonable to keep the blue-white-red

colorscale.

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.

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 (and combine it with Fig. 6) because it adds valueable information to the chapter 3.4.

3   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 "$1/8° \times 1/16°$" → space between "$\times$" 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" → "$1.1° \times 1.1°$", 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

the '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 $AOD_{nitrate}$ further inland over North America." → "that sea-salt is transported further inland over North

C7

America und impacts $AOD_{nitrate}$ 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 $1 < RNS < 30$").