

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

Anonymous Referee #2

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This is a well-written paper that highlights an interesting topic that I had not previously thought about, but makes a lot of sense conceptually and could have important implications for anthropogenic aerosol radiative forcing on a global scale. I have one comment which questions some of the methods used in the experiment, which I am not expecting the authors to address in this paper but could be the subject of further discussion or future work. Beyond that, I just have a few minor comments I think the authors should

My main comment is that the main purpose of this paper is to investigate the sensitivity of the nitrate aerosol radiative forcing to the process that redistributes it from fine modes when associated with ammonium to coarse modes when associated with sodium. The authors have tested this by turning off sea salt emissions in the sensitivity simulation and seeing how this changes nitrate aerosol size distribution, which

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is perfectly valid but does limit what they can investigate in some ways because removing sea salt emissions will have many other changes to total aerosol AOD, CCN and aerosol-radiation/aerosol-cloud interactions that go beyond the redistribution effect they are investigating. I would have thought a better experiment would have been to turn off the association between NO_3 and Na in MOSAIC in the sensitivity simulation, and have sea-salt emissions in both, thereby having a “clean” experiment where the only change is to the exact process you are investigating. A key benefit of this approach is that you would then be able to test the impact of this process on both total aerosol AOD/DRF (not just the NO component), and how it effects the aerosol-cloud interactions (ACI).

On both a local and global scale, my gut says that the influence of the redistribution effect on (ACI), particularly the Twomey or first-indirect effect, would be greater than the DRF. I would expect shifting nitrate from subnitrate modes to coarse modes would slightly increase the number of low-supersaturation CCN, whilst massively decreasing the number of high-supersaturation CCN. This could in turn have a large impact on the activated cloud droplet number, cloud optical depth and therefore climate. Unfortunately, the current methodology cannot address this as any impact from the redistribution effect would be likely overwhelmed by the lack of sea salt aerosol in the sensitivity simulation. This is a shame, as without investigating the aerosol-cloud interactions we are only seeing part of the impact of this process on the climate.

As I said above, I'm not expecting the authors to do reruns with this change as it is a substantial amount of work and, to my knowledge, their current paper is already the first I know of to look into this problem in detail and already contributes significantly to the field. But I would be interested in hearing their response and any follow up studies that look into this further – this discussion could be included in the revised paper.

Minor comments: Ln 40-41: Under what kinds of emission scenarios are we expecting the aerosol radiative cooling to increase by this much?

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Ln 76: Please explain acronym for HOPE-Melpitz campaign Ln 82: “which represent continental period and marine period” Is this based from back trajectories? Please explain better.

Section 2.2 Model Description: this section is missing some important information. Firstly, I couldn't find anywhere a description of how the MOSAIC mechanism describes the chemical uptake of HNO₃ onto aerosol via NH₄⁺ or Na⁺ channels. A short (one-two sentences) here describing that process and how it is parameterised in MOSAIC would be welcome.

Please can you add what meteorology data you are using to drive the model and if you are nudging, or if it freerunning (with or without feedbacks). Some of this information is in the supplement.

Finally, are you running with N₂O₅ heterogeneous chemistry? This is relevant for a point later on in the paper, plus in WRF-Chem the Bertram and Thornton (2009) parameterisation does have a channel for uptake via Cl⁻, so provides another mechanism for nitrate to be taken up to course modes that is modulated by the presence of sea salt (Archer-Nicholls et al., 2014).

Bertram, T. H., & Thornton, J. A. (2009). Toward a general parameterization of N₂O₅ reactivity on aqueous particles: the competing effects of particle liquid water, nitrate and chloride. *Atmospheric Chemistry and Physics*, 9(4), 15181–15214. <https://doi.org/doi:10.5194/acp-9-8351-2009>

Archer-Nicholls, S., Lowe, D., Utembe, S., Allan, J., Zaveri, R. A., Fast, J. D., et al. (2014). Gaseous chemistry and aerosol mechanism developments for version 3.5.1 of the online regional model, WRF-Chem. *Geoscientific Model Development*, 7, 2557–2579. <https://doi.org/10.5194/gmd-7-2557-2014>

Ln 110: Is the version of MOSAIC being used with or without aqueous chemistry?

Ln 137: Please include figure of outer and inner domains, here or in supplement.

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Ln 249: In terms of lifetime of nitrate radical (i.e. gas phase NO_3), I would have thought this has more to do with the uptake of N_2O_5 to aerosol enhanced by Cl^- rather than the HNO_3 process you discuss in this paper. I'm not sure if it is quite relevant with the discussion here, which is more about the lifetime of aerosol NO_3^- when it is in different aerosol size modes. Please clarify.

Ln 336-347: Would it be possible to show a map of RNS over Europe and North America from the WRF-Chem model for comparison? You've only done this for the global EMAC model, which by the sounds of things has lower values for RNS over most of the US compared to WRF-Chem (essentially nowhere in North America has $\text{RNS} > 30$ in the EMAC model, whereas this regime dominates continental North America in the WRF-Chem simulations). The figures currently given make it hard to follow the logic in this paragraph. In general, I think this section can be improved by outlining the conclusions that can be drawn from the evidence given, rather than speculating on what we might expect the effects to be if sea salt is transported further inland.

Ln 348-350: Would it be possible to plot a first-order estimate of what the change to nitrate AOD would be from this effect, using a function based on the relation between RNS and $[\text{NO}_3^-]$ derived from the WRF-Chem model simulations?

Ln 364: “(~90%)” – what is this referring to exactly?

Ln 390-394: This last section is confusing what you can derive from the results of this study (modulation to nitrate DRF from sea-salt aerosol) from that speculated from other related chemical interactions (sulfur, dust etc.). Please rewrite to clearly separate the conclusions drawn from your results from speculations/highlight topics for future research.

Figures:

Please promote Fig S1 into the main paper, I think it really clearly shows how the redistribution effect changes the aerosol chemical size distribution.

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Fig 9. Please can you include a subplot with this showing [NO₃-]? You need this with RNS to get an understanding of where the redistribution impact is likely to be strongest.

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