Authors' response to comments made by anonymous reviewer #2:

Summary

The manuscript titled "Impact of mineral dust on the global nitrate aerosol direct and indirect radiative effect" by Milousis et al. investigated the radiative effects of nitrate on dust by using a climate model, including the aerosol-radiation interactions and aerosol-cloud interactions. Nitrate chemistry on dust is implemented in the EMAC model, simulations were conducted based on the base case and several sensitivity simulations. In general, the logic of the study is explicit and the organization of the manuscript structure is clear. However, the study lacks of necessary evaluation of the simulation results and thus the results could be subject to high uncertainties.

We would like to thank the reviewer for his/her thoughtful review and positive response. Below is a point-by-point response (in black) to his/her comments (in blue).

Major Comments

1. *There is no comparison between model results and observations, e.g. mass concentrations of nitrate, dust (PM10), aerosol number concentrations. There are plenty of observational datasets or literature values available. The lacking of constraints from observational data will reduce the credibility of model simulations.*

We agree with the reviewer that including comparisons with observational data to demonstrate the ability of the model to provide realistic estimates of both aerosol concentrations and cloud droplet numbers will help to increase the credibility of the study's findings. For this reason, we have now included in the supplementary material a comparison of the model results for surface mass concentrations of PM_{10} aerosols with observations from measurement networks in the polluted Northern Hemisphere (EANET, EMEP & IMPROVE). In addition, we have also included a comparison between the CDNCs simulated by the model and those measured in a variety of regions across the world (continental, polluted and clean marine) over different time periods and altitudes, as found in Karydis et al., (2017) and all relevant references therein. The reader is made aware of this content at the end of Section 2.1 in the revised version.

2. *Figure 2 & Line 371 – 377: In Figure 2v and 2vi, the TOA SW REari of coarse nitrate is much stronger than that of fine nitrate, it seems unreasonable as fine nitrate dominates the total nitrate, especially in East Asia.*

This is an excellent point, which helped us discover an error in the simulation where HNO_3 should only condense on the fine mode (i.e., the simulations where the coarse mode was excluded). The error came from the thermodynamic calculations, where only negligible amounts of HNO_3 actually condensed on the fine mode, resulting in unrealistically low fine nitrate concentrations and thus such weak estimates of the radiative effect. This error affected the contribution of the fine and coarse modes to the direct radiative estimate of total nitrate (namely $F_{fN,ari}$ & $F_{cN,ari}$) and not the estimate of the total nitrate aerosol itself ($F_{N,ari}$) in Section 2.3.1. This is because only the

quantity $F_{3,ari}$ (calculated taking into account all aerosol components except coarse NO_3^-) was incorrectly calculated. Therefore, this error was not transferred to the calculation of the indirect radiative estimate, since only the quantity $F_{N,ari}$ was used for this (Section 2.3.2). We have now corrected this error by ensuring that for simulations where the coarse mode is removed from the aerosol load, the thermodynamic calculations for HNO_3 condensation are performed correctly and the condensed HNO_3 is only transferred to the fine mode. For this reason, we have performed 5 new simulations (1 for each sensitivity case) where these conditions apply. As a result, Figure 2 and Table 2 have been updated with the correct results for the fine and coarse mode estimates for the direct radiative effect. In addition, Sections 3.1 and 3.2 have been thoroughly revised to incorporate the new results.

3. *Figure 4: the kappa values of fine aerosol over the continents are mostly lower than 0.04 (iii), which are incorrect. Even considering the mixing between dust and anthropogenic emissions, the hygroscopicity of aerosols couldn't be so weak.*

While the kappa values of the fine aerosol population appear to be low, particularly over the dust belt zone, this is largely due to the higher proportion of insoluble fine aerosols present there. This is also observed over other regions with similarly low fine aerosol hygroscopicity (South Africa, South America and Western U.S). Furthermore, the estimates of aerosol kappa values at 940 hPa are broadly in agreement with the findings of Pringle et al., (2010). We have included the model estimates for the global insoluble fractions of the fine and coarse aerosol populations in the revised supplementary material. The reader is referred to this in the relevant part of Section 5.1 in the revised manuscript.

4. *Line 195 – 197: Na+, K+, Ca2+ and Mg2+ constituted 100% of bulk dust? How are the anions treated?*

No, this is not the case. The composition of the emitted mineral dust consists of a bulk component, which accounts for 94% of the emitted flux, and the remaining 6% represents the mass fractions of the mineral cations. No anions are considered to be explicitly emitted as part of the emitted dust flux.

5. *Line 222 – 223: How is the delinquencies of salts treated in the model under different relative humidity?*

In our model, the deliquescence of salts under different relative humidities is treated according to the Mutual Deliquescence Relative Humidity (MDRH) approach of Wexler and Seinfeld (1991), as described in Fountoukis and Nenes (2007). More specifically, each individual salt has a certain threshold, the DRH, above which its phase transition from solid to liquid occurs. However, in the presence of a multicomponent mixture, it is the MDRH that determines the humidity value above which all salts in the mixture are considered to be saturated. The MDRH is below the DRH of all the pure solids in the mixture. As the RH over a wet particle decreases, the aerosol may not crystallize below the MDRH but instead remain in a state where it consists of an aqueous solution that is supersaturated with dissolved salts. This state is called metastable and is the state considered in our study by the ISORROPIA-lite thermodynamic model (Kakavas et al., 2022; Milousis et al.,

2024). This information has been added in the revised version of the manuscript in Section 2.2 right after the deliquescence chemical reactions.

6. Section 4: Why the radiative effects from Aerosol-Cloud Interactions are not separated for fine and coarse nitrate?

This is a valid question. As explained at the beginning of Section 2.3.2 on the REaci calculation methodology, we estimate it in this way because it is essential to include feedbacks from different climatological conditions. In particular, since climatology plays a crucial role in aerosol-cloud interactions, the simulation of a "fine-only NO_3^- atmosphere", as done for the REari calculations, would produce a climatological scenario that would lead to inaccurate estimates of the feedback radiative effect of nitrate aerosol. This is because coarse-mode NO_3^- is strongly associated with cations in mineral dust particles (Karydis et al., 2016), making them quite effective as CCN (Karydis et al., 2017). Separating of the direct radiative effect between fine and coarse nitrate is a simpler task that provides realistic results because of two conditions. First, not only are the aerosol-cloud interactions switched off (by not considering any specific parameterization for aerosol activation besides the cloud cover prognostic equations, as described at the end of Section 2.3.1), but also any potential uncertainties due to different climatological conditions are eliminated in this case, as explained in Section 2.3.1.

References

Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+–Ca^{2+}–Mg^{2+}–NH_4^+–Na^+–SO_4^{2-}–NO_3^-–Cl^-–H_2O aerosols, Atmos. Chem. Phys., 7, 4639–4659, https://doi.org/10.5194/acp-7-4639-2007 , 2007.

Kakavas, S., Pandis, S. N., and Nenes, A.: ISORROPIA-Lite: A Comprehensive Atmospheric Aerosol Thermodynamics Module for Earth System Models, Tellus Series B-Chemical and Physical Meteorology, 74(1), 1-23, https://doi.org/10.16993/tellusb.33 , 2022.

Karydis, V. A., Tsimpidi, A. P., Pozzer, A., Astitha, M., and Lelieveld, J.: Effects of mineral dust on global atmospheric nitrate concentrations, Atmospheric Chemistry and Physics, 16(3), 1491-1509, https://doi.org/10.5194/acp-16-1491-2016 , 2016.

Karydis, V. A., Tsimpidi, A. P., Bacer, S., Pozzer, A., Nenes, A., and Lelieveld, J.: Global impact of mineral dust on cloud droplet number concentration, Atmospheric Chemistry and Physics, 17(9), 5601-5621, https://doi.org/10.5194/acp-17-5601-2017 , 2017.

Milousis, A., Tsimpidi, A. P., Tost, H., Pandis, S. N., Nenes, A., Kiendler-Scharr, A., and Karydis, V. A.: Implementation of the ISORROPIA-lite aerosol thermodynamics model into the EMAC chemistry climate model (based on MESSy v2.55): implications for aerosol composition and acidity, Geoscientific Model Development, 17(3), 1111-1131, https://doi.org/10.5194/gmd-17-1111-2024 , 2024.

Pringle, K. J., Tost, H., Pozzer, A., Pöschl, U., and Lelieveld, J.: Global distribution of the effective aerosol hygroscopicity parameter for CCN activation, Atmos. Chem. Phys., 10, 5241–5255, https://doi.org/10.5194/acp-10-5241-2010 , 2010.

Wexler, A. S., & Seinfeld, J. H.: Second-generation inorganic aerosol model. *Atmospheric Environment. Part A. General Topics*, *25*(12), 2731-2748, https://doi.org/10.1016/0960-1686(91)90203-J, 1991.