

Referee #2

The authors investigated global major inorganic aerosols and the effect of various cations in dust and sea salt on the formation of aerosol nitrate using the ECHAM5/MESSy Atmospheric Chemistry model (EMAC). They further designed a suite of sensitivity experiments to demonstrate systematically the impact of the strength and chemical composition of dust emission in atmospheric nitrate formation. This is an interesting and valuable study. I recommend publishing the paper in ACP after the authors make some minor modifications.

We would like to thank Dr. Huisheng Bian for her positive response and for raising important issues. Below is a point by point response to her comments.

General Remarks:

- 1. The terminology of “mineral dust” used in the title and discussion is not appropriate. The authors investigate the importance of four cations (i.e. Ca^{2+} , Mg^{2+} , K^+ , Na^+) in the formation of nitrate and other atmospheric inorganic aerosols. However, these four cations, according to the description of model setup, come from not only mineral dust emission but also sea spray aerosols, and the latter is particularly important for Na^+ and Mg^{2+} . It may be worthwhile to introduce an additional sensitivity simulation by turning off dust-only (or sea salt-only) cations and comparing it with the base case simulation to identify contributions from the corresponding aerosol.*

It is true that in our model setup sea salt include the four mineral cations as well (Ca^{2+} , Mg^{2+} , K^+ , Na^+). However, sea salt emissions and properties (chemical composition, size distribution, etc.) never changed through our sensitivity tests. We only changed the properties of mineral dust. For instance, in simulation case 2 (Table 5 of the manuscript) we have turned off the above four cations only from dust emissions (and not sea salt). Therefore, with this sensitivity simulation (which is identical with the one that the referee proposed) we managed to identify the effect of mineral aerosols on nitrate formation. In order to avoid any confusion we have renamed this simulation case to “Chemically inert dust” (as the first referee also proposed) and we clearly state in the revised manuscript that we remove these cations only from mineral dust aerosols and not from sea salt.

- 2. The evaluation over North America (NA) using IMPROVE measurement needs to be revisited. IMPROVE measures fine mode nitrate only (i.e. 2.5 μm in diameter, see <http://vista.cira.colostate.edu/DatawareHouse/IMPROVE/Data/AEROSOL/Help/IMPROVEVarTable.txt>). However, North America, particularly western US, has noticeable coarse mode nitrate (i.e. various nitrate salts) associated with the discussed cations. Cautions should be taken when comparing the model simulation with the IMPROVE measurement. Another useful surface measurement network over NA is CASTNET, which provides measurements of surface nitrate, ammonium, and sulfate.*

We agree with the reviewer that the use of the CASTNET network is more appropriate for this study. Therefore, in the revised manuscript we have removed the comparison against the IMPROVE network and we now present the evaluation of our model predictions for aerosol nitrate, sulfate, chloride, sodium, calcium, magnesium, and potassium concentrations against the CASTNET network measurements.

Specific comments:

1. *Page 11526 line 13-14 (abstract): How do the authors know the updated model improves nitrate predictions over remote areas? I do not find this discussion in the paper.*

This is correct. We have not made any evaluation of the previous set up of the model to justify this. Therefore in the revised manuscript we have removed this sentence from the abstract.

2. *Page 11527 line 19-23: Logically, “in polluted regions” in line 21 should be moved to the sentence above.*

Done

3. *Page 11530 line 13-15: Add “compiled from literatures” after “... the main deserts”.*

Done

4. *Page 11530 line 22: Please clarify “lower and middle atmospheric”, such as from troposphere to stratosphere (or mesosphere), or from surface to how many km.*

In the revised manuscript it has been clarified that EMAC includes sub-models describing atmospheric processes from the troposphere to stratosphere.

5. *Page 11531 line 8: What is the difference between MECCA and MESSy2?*

MESSy2 is the Modular Earth Submodel System that links several submodels with a core base model. MECCA is one of the submodels that describes the gas-phase chemistry. A more detailed description of MESSy2 has been added in the revised manuscript.

6. *Page 11531 line 15: How about wet deposition since the authors have described dry deposition and sedimentation?*

The wet deposition of trace gases and aerosol particles is calculated within the SCAV sub-model (Tost et al., 2006). This information has been added to the text.

7. *Page 11532 line 4-6: Are these salts treated as independent tracers in dynamic transport and dry and wet depositions?*

No, these compounds are used only by ISORROPIA II for the calculation of the gas/liquid/solid equilibrium partitioning of the inorganic ions.

8. *Page 11532 line 19: Please elaborate on “specific input fields”.*

The most important input fields for the soil properties are the geographical location of the dust sources, the clay fraction of the soils, the rooting depth, and the monthly vegetation area index (sum of leaf and stem area index). This clarification has been made to the revised manuscript.

9. *Page 11533 line 19-20: What types of NH₃ are included in “natural emission”?*

The natural emissions of NH₃ include excreta from domestic animals, wild animals, synthetic nitrogen fertilizers, oceans, biomass burning, and emissions from soils under natural vegetation. This information has been added to the text.

10. *Page 11533 line 23-24: How about SO₂ emission from volcanic eruption?*

Both eruptive and non-eruptive volcanic degassing emissions of SO₂ are based on the AEROCOM data set (Dentener et al., 2006). This clarification has been made in the revised manuscript.

11. *Page 11533 line 25-26: Check the unit of emissions. Should it be TgNyr-1 or TgSyr-1?*

The units are Tg y⁻¹ and not TgN yr⁻¹ or TgS yr⁻¹.

12. *Page 11536 line 20: Change “inter-annual average” to “multi-year average”.*

Done

13. *Page 11540 line 9-10: I am not convinced of this sentence with the reasons given by the authors. For example, missing a consideration of water soluble organic acids may help the partitioning favor nitrate aerosol since lab experiments indicated that organic acids can accelerate re-cycle nitrate aerosol back to gas phase nitric acid.*

This is a valid point made by the reviewer. In the revised manuscript we have removed any statement that is based on vague assumptions and we provide the statistical evaluation of the modeled total HNO₃ (gas+aerosol) to confirm that the nitrate aerosol overestimation by the model is due to the overprediction of total HNO₃ and not due to errors in its phase partitioning. This is also in accordance to the reviewer’s next comment.

14. *Page 11540 line 12-13: The authors can confirm this by comparing the model HNO₃ with measurement.*

We have compare our model results against measurements of total HNO₃ concentrations measured by the EMEP network and we have confirmed that the model significantly overpredicts total nitrate over Europe with MB=2.29 µg m⁻³. This information has been added to the revised manuscript.

15. Page 11545 line 11-13: Please elaborate on how the model treats equilibrium in two modes. How does the model divide the two modes?

The aerosol size distribution is described by 7 interacting lognormal modes (4 hydrophilic and 3 hydrophobic modes). The 4 hydrophilic modes are arranged to cover the aerosol size spectrum (nucleation, Aitken, accumulation and coarse). The 3 hydrophobic modes have the same size range, but no hydrophobic nucleation mode is required. Each mode is defined in terms of the number concentration, the number mean radius and the geometric standard deviation (σ) and has a fixed size boundary but a variable mean radius (Pringle et al., 2010).

The assumption of thermodynamic equilibrium is a good approximation for fine mode aerosols which can reach equilibrium very fast. However, the equilibrium timescale for large particles is typically larger than the timestep of the model (Meng and Seinfeld, 1996). To account for kinetic limitations the process of gas/aerosol partitioning is calculated in two stages (Pringle et al., 2010). In the first stage the amount of the gas phase species that are able to kinetically condense onto the aerosol phase within the model timestep is calculated assuming diffusion limited condensation (Vignati et al., 2004). In the second stage ISORROPIA-II re-distributes the mass between the gas and the aerosol phase assuming instant equilibrium between the two phases.

This discussion has been added in sections 2.1 and 2.2 of the revised manuscript.

16. Page 11547 line 17-18: Why is atmospheric dust load not half when emission is cut to half? Are the nitrate salts treated as independent tracers outside ISORROPIA-II?

When dust emissions cut to half the soluble fraction of dust increased almost by 2%. This increase in the solubility of dust resulted in its slightly higher atmospheric removal through wet deposition compared to the base case simulation leading to a non-linear response of mineral dust tropospheric burden to the applied emission change.

17. Page 11548 line 1: It would be good if the authors could mark the discussed various deserts on a map.

We have added the figure 1 in the revised manuscript which depicts a map with the location of the discussed desserts in this study.

18. Page 11549 line 27: Change “change” to “reduction”.

Done

19. Page 11568 Figure 2: Suggest changing (b) to nitrate aerosol and (c) to fraction of fine mode nitrate to provide more information.

We have adopted the reviewer's suggestion and we have replaced the figures 2b and 2c (Figures 3b and 3c in the revised manuscript) with the nitrate aerosol and the fraction of fine mode aerosol nitrate, respectively.

20. Page 11571 Figure 5: Add experiment name after "A positive change corresponds to a decrease".

Done

Technique corrections:

1. Page 11529 line 27: Change "have include" to "have included".

Done

2. Page 11531 line 1: Please add the types of observation in "observations and satellite measurements".

The EMAC model has been extensively described and evaluated against in situ observations and satellite measurements that include filter based particulate matter concentrations, aerosol optical depth, acid deposition, gas phase mixing ratios, and meteorological parameters, and can be applied on a range of spatial resolutions. The above sentence has been added in the revised manuscript.

3. Page 11546 line 2-3: It is better to have this sentence the same as in Figure 6.

Done

4. Page 11562 Table 1: Change "Dust" to a term such as "other", "remnant", etc.

Done