Anonymous Referee #1

General comments

1. *The paper addresses relevant scientific questions within the scope of ACP. The authors do not clearly support to present novel concepts, ideas, tools, and data, but they analyze an interesting topic, i.e. the effect of desert dust on inorganics on the global scale, focusing on aerosol nitrate. The overall presentation is clear; some issues in the structure are pointed out. The last paragraph of the Introduction fails to support the motivation and goals pursued in the Results. Although results are sufficient to support the conclusions, more interpretations are needed for some findings. All of the above comments (and some more) are specifically described below and in the attached file.*

We would like to thank the referee for the thoughtful and detailed review and also for all suggestions to restructure the manuscript in order to better present our results. Below is a point by point response on the comments and suggestions.

Specific comments

2. *Abstract: the abstract should be revised after all comments are taken into account, so that it serves as a concise and complete summary of the article. Indicative rewording is shown in the attached file.*

The abstract has been revised accordingly.

3. *Introduction: Each paragraph should have a clear and concise concept that serves to cover a specific aspect dealt in this work. Also, paragraphs should (in)-directly try revealing the new/original contribution of the current study. Apart from these general statements, specific comments are given in the attached file. Although a search in previous relevant studies was redone, I still find some missing (cf. attached file). After going through the whole text, I realized that the last introductory paragraph presented the work in a simplistic way. In other words, the study analyzes the physics and chemistry of dust on a global scale, but it is primarily presented as an improvement in modeling. I would suggest to rewrite this paragraph, so that it better supports the idea behind the paper, its motivation, goals and findings.*

The introduction has been revised based on the comments of the reviewer. Furthermore, the last paragraph of the introduction has been rewritten in order to illustrate that the main goal of this study is to assess the effect of naturally emitted dust particles on global nitrate aerosol concentrations and size distributions and to present the modeling tools and methods that have been used to achieve this goal.

3.1 page 11527, line 15: what all these studies have found?

A statement has been added to the manuscript reporting the findings of these studies regarding the tropospheric burden of aerosol nitrate.

3.2 page 11528, line 9: although the list is expanded, I still locate omissions

More references with respect to prediction improvements when sea salt and dust chemistry is incorporated in model applications have been added to the manuscript following the reviewer's recommendations.

3.3 page 11528, line 14: Instead of presenting the findings of indicative studies on this issue, place sequentially (aerosol improvements when sea-salt and/or dust particles are incorporated), you should present their common findings, i.e. please make a sum of their results, to be presented as a whole and not separately. In case you want to stress out some findings, this should be done only for those directly linked to your study/results.

In this paragraph we present the findings of several studies regarding the effect of sea salt and soil particles on aerosol thermodynamics. However, since not all of these studies have resulted in similar conclusions, we cannot present their results as a whole. Instead, in the revised manuscript we summarize the above studies in proportion to their findings and we discuss their results.

3.4 page 11528, line 23: please, check if more of the above studies (including my suggestions) have used the dynamic calculation of mass transfer (fine and/or coarse mode). if so, again make an overall statement of their findings.

An overall statement presenting the findings of all the studies that have used the hybrid approach has been added to the text.

3.5 page 11529, line 20: you begin and end this paragraph with a similar statement. why not give the main point of the paragraph and then analyse it further, using the common findings of all relevant publications?

The goal of this paragraph is to point out that there are only a few global model studies that account for the dust aerosol chemistry. However, even these studies have made some bulk assumptions about the mineral dust properties and have not assessed the impact of dust on nitrate aerosol formation. In this context, the first part of this paragraph points out the fact that most of the global models do not include dust aerosol chemistry at all while the last part of the paragraph highlights the disadvantages of the few existing studies that include dust aerosol chemistry.

3.6 page 11529, line 21: I am confused here: the studies in parenthesis lack realistic treatment, right? each of the following studies being analysed below (eg feng and penner) are: 1. additional ones (lacking realistic treatm.)? 2. should be included in the parenthensis? also, why you analyze only those and not the initially mentioned?

The studies in parenthesis lack a realistic treatment of dust. On the other hand, the following studies do include interactions of mineral dust with nitric acid. This is also the reason why we focus only on the findings of these studies and not on the initial ones. To avoid any confusion we have clarified this in the revised text.

3.7 page 11530, line 13: what do you mean?

We have applied a different chemical composition of dust for each of the main deserts of the world based on the cited literature. In order to avoid any confusion we have rephrased the sentence to: "Chemical compositions of the emitted dust particles compiled from the literature are adopted for the main deserts to study the chemical interaction among crustal and inorganic species".

3.8 page 11529, line 16: explain more clearly the test: on the scheme or on the type of texture?

We have used an alternative emission scheme which uses a uniform size distribution for all types of soil textures. To clarify this we have replaced the "dust emission scheme based on the soil texture" with the "size distribution of the soil particles".

4. Sect. 2: comments are shown in the attached file.

4.1 page 11531, line 27: Did you perform the coupling between emac and isorropia II, in the frame of this study? if so, please state this clearly.

No we did not. ISORROPIA II was already part of EMAC (see Pringle et al. 2010). In this study we used the model configuration that includes the crustal species and we have assigned emissions to them using the online dust emission scheme and the chemical composition of dust from each desert.

5. Sect. 3: some kind of restructuring and elaboration is needed (cf. attached file).

5.1 page 11534, line 4: since the main target of this study is mineral dust, I would suggest to structure the results in a balanced way with respect to that. in specific, the

chemical constituents of dust, as shown in table 1, should be subsections of 3.1. then, 3.2 could titled like 'rest inorganic aerosol species'.

Following the reviewer's recommendation we have restructured section 3 (Section 4 in the revised manuscript) by using three subsections. Since this study focuses on the effect of dust chemistry on nitrate aerosol concentrations we have grouped the discussion in this section as follows: In section 4.1 we discuss the results for mineral dust and its chemical constituents, section 4.2 refers to particulate nitrate and gaseous nitric acid concentrations, and in section 4.3 presents the results for the rest of the inorganic aerosols.

5.2 page 11535, line 18: it could be nice to see a general comment with respect to calcium, i.e. the spatial distribution of potassium is similar than that of calcium due to their common origin (dust), except from .... . please check this option also with respect to the other subsections, i.e. it would be easier for the reader to be guided on common findings and reasons for differentiations.

We have added general statements in all subsections that compare the constituents to each other, making the identification of the similarities and differentiations between the components easier for the reader.

5.3 page 11536, line 25: please reverse order

Done.

5.4 page 11537, line 11-14: I do not agree with this statement: assuming equilibrium allows for a instantaneous condensation of all available sulfuric acid upon solid particles, thus more no3 will remain in the gaseous state. on the other hand, the dynamic solution of the mass transfer equations means a gradual condensation of gases and will ensure more particle surface available for nitrate condensation, too.

We agree with the reviewer that this may be the case when the sulfuric acid concentrations are high and there is not enough ammonia to completely neutralize the sulfate aerosols (which will lead the sulfuric acid to the fine mode). Therefore, we have added the following statements in the part of our discussion that refers to the anthropogenically affected areas: "Over these areas, where sulfuric acid is high, bulk equilibrium assumption can result in underprediction of coarse nitrate since it allows for instantaneous condensation of all the available sulfuric in the aerosol phase, leaving more nitrate in the gaseous state. On the other hand, a dynamic solution of the mass transfer equations will result in a gradual condensation of gases and will ensure more particle

surface available for nitrate condensation." However, over Central Africa sulfur concentrations are low and nitric acid is in excess due to its high emissions from biomass burning. In this case, the presence of sulfate in the aerosol (especially in the coarse mode) is limited and the equilibrium assumption will lead most of the nitric acid in the coarse aerosols. A dynamic approach will predict lower nitrate aerosol concentrations compared to the equilibrium approach since the time step of the model is not enough for the gradual condensation of all the available nitric acid. To emphasize that this statement is valid only under certain conditions we have rephrase it to: "Taken into account that sulfuric acid concentrations over Central Africa are low, nitric acid (which is in excess) is practically the only available acid in the atmosphere to react with the mineral cations. In this case, the assumption of thermodynamic equilibrium in the coarse mode may result in an overprediction of coarse aerosol nitrate."

5.5 page 11537, line 15-16: since this is not performed in the current simulations (as a base-case or sensitivity), a comment on why would be appreciate.

The reason we didn't use the hybrid approach is because of its computational cost, especially for global scale calculations. Instead, in order to account for the kinetic limitations of the gas/aerosol partitioning in the coarse mode, we used in the equilibrium calculations only the fraction of the gas that could kinetically condense within the time step of the model. The following discussion has been added to the text: "However, the additional calculations required for the dynamic solution of the mass transfer equations adds significantly to the computational overhead of the model. In this study, the kinetic limitations of the gas/aerosol partitioning in the coarse mode are considered by using only the fraction of the gas that could kinetically condense within the time step of the model, in the equilibrium calculations." Furthermore, the method used in this study to account for non-equilibrium conditions is now described in section 2.2 of the revised manuscript.

6. *Sect. 4 & 5: again, ideas for a different structure, as well some questions to be answered (cf. attached file).*

6.1 page 11538, line 22: again, this section comes to disorientate the reader: although the focus of this work is mineral dust and its interaction with nitric substances, model evaluation is extended enough, focuses on different continents and loses contact with the main target. i would suggest to move it in front (as section 3), condense it as much as possible, and more importantly structure each subsection, as 'dust related' (including nitrates) and 'rest' chemical constituents of aerosols.

Following the reviewer's recommendation we have moved this section before the model results and we have restructured it in 3 subsections (mineral dust, nitrate, and rest inorganic aerosol components) based on the aerosol species and not the continents.

6.2 page 11540, line 22-23: which specific characteristics of the representation over the sahara contribute to the good estimations, when compared to the representation over other deserts by the current applications?

We mainly refer to the total mineral dust emissions and the chemical composition of dust used for the estimation of the emissions of the reactive mineral components. This clarification has been added to the text. However, EMEP network measurement stations are affected from dust originating from the Sahara desert. Therefore, we cannot make any conclusions or any comparisons with the rest of the deserts.

6.3 page 11541, line 1: or USA? please be consistent throughout text.

We now use "North America" throughout the evaluation discussion.

6.4 page 11541, line 2-3: do you mean that the model performs the best over N. America, when compare to model performance over the other continents (europe/4.1 etc)?

Yes, this is what we mean. We have clarified this in the revised manuscript.

6.5 page 11542, line 17: couldn't this be erased?

Done.

6.6 page 11542, line 20: please rephrase

The phrase has been rewritten as: "Therefore, the impact of mineral dust on nitrate aerosol formation over Asia calculated by this study is probably underestimated as well."

6.7 page 11542, line 24: couldn't this be erased?

Done.

6.8 page 11543, line 15: is there any particular reason, you dont start section 5 by analyzing first the impact on all inorganic species, and then proceed to the effects on nitrates (5.1, and 5.3)?

We have change the order of the subsections and in the revised manuscript we start with the effect on inorganic aerosol concentrations.

6.9 page 11543, line 16: also i do not understand the paragraph structure of sect. 5.2 (it should not be just that you comment on fig and table 5): unless the first paragraph has a specific concept, different than the next, then an idea is to split 5.2 content per species.

Following the reviewer's recommendation we have change the structure of section 5.2 (5.1 in the revised manuscript) and we have split it in two subsections: 5.1.1 Effects on nitrate aerosols and 5.1.2 Effects on the rest inorganic aerosol components.

6.10 page 11543, line 16: as indicated in my other comments, i think a more appropriate terminology should be used for each scenario. please, reassess.

Following the reviewer's recommendation we have rephrased the statement to: "In the sensitivity simulation in which mineral dust is assumed to be chemically inert,"

7. Sect. 6: although interesting results are presented one-by-one, they are not summarized and some need further explanation (cf. attached file).

7.1 page 11545, line 21: to my view, here you describe your results one by one, without trying to synthesize them somehow. thus, it is advised to do so in sect. 7.

The revised manuscript includes two paragraphs in section 7 (Summary and conclusions) where we synthesize the results from our sensitivity tests and we discuss the relative importance of the examined parameters both on the calculated tropospheric burden of aerosol nitrate and on the nitrate aerosol concentrations predicted at various locations in the world.

7.2 page 11546, line 15: i would like to see some reasoning for: the reduction of emissions and the changes per region.

As described in the text, the reduction in emissions is a result of the substantially different soil particle size distributions and emitted size distributions which influence the calculated threshold friction velocity and thus the dust aerosol emission fluxes. The sensitivity simulation produces weaker emissions than the base case, mostly due to differences in the Asian and South American deserts (two to three times lower emissions) and to a lesser extent in Saharan, Arabian and Australian deserts. These changes in dust

emissions drive the impact on nitrate concentration predicted by the sensitivity simulation in each of the above regions. This information has been added to the revised manuscript.

7.3 page 11546, line 21: America vs USA vs US: please be consistent throughout text

In the revised manuscript we use "USA" when we refer to the country (mainly on sections 5 and 6) and North America when we refer to the continent (mainly in the evaluation section).

7.4 page 11547, line 6: why?

The fraction of the individual mineral components to total mineral dust assumed in the sensitivity simulation is lower in most of the deserts compared to the base case applied chemical compositions which result in their reduced emissions affecting their calculated tropospheric burden. This discussion has been added to the text.

7.5 page 11547, line 8: why?

Nitrate aerosol tropospheric burden is reduced since the mineral cations are reduced substantially in the sensitivity simulation. We have pointed out in the revised text that the tropospheric burden of mineral components (Na^{+}, Ca^{+2}, K^{+}, and Mg^{+2}) reduces by 17%, 40%, 37%, and 48%, respectively in the sensitivity simulation which results in a decrease of nitrate aerosol tropospheric burden by 16%.

7.6 page 11548, line 11: you dont give your values, so that the clear connection to the theory above is shown.

The sulfate to nitrate molar ratios and the RH values over the areas discussed in this section are now given in the revised manuscript.

*7.7 page 11548, line 15:*give values from your results

The sulfate to nitrate molar ratios and the RH values over the Central Asian deserts and the Atacama Desert are now given in the revised manuscript.

*7.8 page 11548, line 17:*any reasons for these findings? i would rephrase like: " the largest avs increase (0.15...) is calculated .... China (reason i.e. molar ratio...RH values).

The sulfate to nitrate molar ratios and the RH values over northeastern China and the Congo Basin are now given in the revised manuscript.

8. Sect. 7: comments for improvement are given in the attached file.

8.1 *page 11548, line 20:*to my view, these are not just conclusions but also a summary. 'Summary and conclusions' is advised as the title for sect. 7.

We have changed the title to 'Summary and conclusions'.

8.2 *page 11549, line 14:*I would expect (not exactly at this point, but in general in the discussion) to see a comment on how the parameters you study with all sens. tests, can be important (or in contrast, insignificant) for a continent vs. the others.

We have added a paragraph at section 7 (Summary and Conclusions) to discuss the relative importance of the examined parameters on nitrate aerosol formation over various locations around the world.

8.3 *page 11550, line 1:*i propose indicative numbers % in parenthesis

The percentage change in the nitrate aerosol tropospheric burden calculated by the sensitivity simulations has been added in the revised text.

8.4 *page 11550, line 6:*again, this paragraph is rather vague. it is advised to use numbers i.e. your results, wherever possible.

This paragraph serves mostly as a synopsis of our work and aims to emphasize the importance of including the thermodynamic interactions of nitrate with mineral cations in global models. Therefore it does not contain any new results that have not been presented before in the conclusion section. However, we have followed the reviewer's suggestion and we have used numbers for presenting our results in the conclusion section wherever possible.

9. Minor comments in some tables and figures are given in the attached file.

All comments and suggestions have been taken into account in the revised manuscript

10. The language is quite fluent and precise. Where appropriate, specific directions are given (cf. attached file).

All suggested corrections and rewording have been implemented in the revised manuscript.