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.

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 _m 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 line19-23: 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 NH_3 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 SO_2 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 HNO_3 (gas+aerosol) to confirm that the nitrate aerosol overestimation by the model is due to the overprediction of total

HNO_3 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 HNO_3 concentrations measured by the EMEP network and we have confirmed that the model significantly overpredicts total nitrate over Europe with MB=2.29 $\mu g\ m^{-3}$. 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

Effects of mineral dust on global atmospheric nitrate concentrations

Karydis V.A.[1*], Tsimpidi A.P.[1], Pozzer A.[1], Astitha M.[2] and Lelieveld J.[1,3]

1 Max Planck Institute for Chemistry, Mainz, 55128, Germany

2 Department of Civil and Environmental Engineering, University of Connecticut, USA

3 Energy, environment and water research center, Cyprus Institute, Nicosia, 1645, Cyprus

*corresponding author e-mail: v.karydis@mpic.de

Abstract

This study ~~provides an~~ assess~~ment of~~ the chemical composition and global aerosol load of the major inorganic aerosol components ~~and determines the effect of mineral dust on their formation,~~ focusing on mineral dust and aerosol nitrate. T~~o account for this effect, t~~he mineral dust aerosol components (i.e., Ca^{2+}, Mg^{2+}, K^+, Na^+) and their emissions are ~~added to~~included in the ECHAM5/MESSy Atmospheric Chemistry model (EMAC). Gas/aerosol partitioning is simulated using the ISORROPIA-II thermodynamic equilibrium model that considers ~~the interactions of~~ K^+-Ca^{2+}-Mg^{2+}-NH_4^+-Na^+-SO_4^{2-}-NO_3^--Cl^--H_2O aerosol components. Emissions of mineral dust ~~aerosol components (K$^+$-Ca^{2+}-Mg^{2+}-Na$^+$)~~ are calculated online by taking into account the soil particle size distribution and chemical composition of different deserts worldwide. ~~The p~~Presence of ~~the~~ metallic ions ~~on the simulated suite of components~~ can substantially affect the nitrate partitioning into the aerosol phase due to thermodynamic interactions. The ~~updated~~ model ~~improved the nitrate predictions over remote areas and~~ simulates highest ~~found that the~~ fine aerosol nitrate concentration ~~is highest~~ over urban and industrialized areas (1-3 μg m^{-3}), while coarse aerosol nitrate is highest close to deserts (1-4 μg m^{-3}). The ~~contribution~~ influence of mineral dust ~~components to~~ on nitrate formation ~~is large in areas with high dust concentrations with impacts that can~~ extends across southern Europe, western USA and northeastern China. The tropospheric burden of aerosol nitrate increases by 44% by considering ~~the~~ interactions of nitrate with mineral dust ~~cations~~. The calculated global average nitrate aerosol concentration near the surface increases by 36% while the coarse and fine mode concentrations of nitrate increase by 53% and 21%, respectively. Other inorganic aerosol components are affected by reactive dust components as well (e.g., the tropospheric burden of chloride increases by 9%, ammonium decreases by 41%, and sulfate increases by 7%). Sensitivity tests

show that nitrate aerosol ~~formation~~ is most sensitive to the chemical composition of the emitted mineral dust, followed by the soil size distribution of dust particles, the magnitude of the mineral dust emissions, and the aerosol state assumption.

1. Introduction

Atmospheric aerosols from natural and anthropogenic sources adversely affect human health and play an important role in changing the Earth's climate. Inorganic particulate nitrate constituents contribute significantly to the total aerosol mass, especially in urban areas and industrialized regions (Putaud et al., 2004; Kerkweg et al., 2007; Henze et al., 2009; Kopacz et al., 2010; Jöckel et al., 2010). Over Europe, particulate nitrate accounts for about 10–20% of the total dry aerosol mass (Putaud et al., 2004). Veefkind et al. (1996) suggested that nitrate is particularly important in the optically active submicron size range, related to its ability to efficiently scatter solar radiation and its potential to affect cloud properties. The Intergovernmental Panel on Climate Change (IPCC) also underscored the important role of nitrate aerosol in climate change (IPCC, 2013). However, there is large uncertainty regarding the global nitrate aerosol load, its regional distribution, and its radiative forcing. In fact, only a limited number of global models have been used to estimate particulate nitrate concentrations and their regional distributions (Adams et al., 1999; Metzger et al., 2002; Liao et al., 2003; Rodriguez and Dabdub, 2004; Feng and Penner, 2007; Pringle et al., 2010; Fairlie et al., 2010; Bellouin et al., 2011; Xu and Penner, 2012; Pozzer et al., 2012; Hauglustaine et al., 2014). In these studies, estimates of the nitrate aerosol tropospheric burden vary by one order of magnitude ranging from 0.13 to 1.85 Tg.

One of the challenges in atmospheric aerosol modeling is to compute the partitioning of semi-volatile nitrate between the gas and aerosol phases. Nitrate aerosols in polluted regions are typically formed when sulfate aerosols are irreversibly neutralized and atmospheric ammonia is present in excess. Therefore, ~~in polluted regions~~ nitrate predominantly occurs in the fine mode, mainly observed in the form of ammonium nitrate at continental sites (TenBrink et al., 1997; Putaud et al., 2010). Many thermodynamic equilibrium models have been developed over the past decades that can accurately describe the formation of ammonium nitrate in the aerosol phase (i.e., AIM of Wexler and Seinfeld, 1991; SCAPE of Kim et al., 1993; EQUISOLV of Jacobson et al., 1996; ISORROPIA of Nenes et al., 1998; GFEMN of Ansari and Pandis, 1999). However, aerosol nitrate is not only associated with ammonium in the fine mode. Coarse mode aerosol nitrate can be produced by adsorption of nitric acid on sea salt (Savoie and Prospero, 1982) and soil (Wolff, 1984) particles. In particular the light-metallic ions of calcium, magnesium, sodium, and potassium can be associated with nitrate and affect its partitioning into the aerosol phase. In order to account for the effect of crustal species on the partitioning of nitrate, mineral cations (i.e., Ca^{2+}, Mg^{2+}, K^{+}) have been added to the suite of components of a few thermodynamic models ~~(~~i.e., SCAPE2 (Kim and Seinfeld, 1995), EQUISOLV II (Jacobson, 1999), EQSAM3 (Metzger and Lelieveld, 2007), ISORROPIA II (Fountoukis and Nenes, 2007).

Several studies in the past have shown that ~~The~~ the simulation of these effects, especially in areas where dust or sea salt comprises a significant portion of total particulate matter, can considerably improve model predictions (Dentener et al., 1996; Gong et al., 2007; Jacobson, 1999; Jacob, 2000; Song and Carmichael, 2001; Moya et al., 2002; Bian and Zender, 2003; Laskin et al., 2005; ~~San Martini et al., 2005;~~ Hodzic et al., 2006; Kallos et al., 2007; ~~Zaveri et al., 2008;~~ Astitha et al., 2008; Athanasopoulou et al., 2008; Fountoukis et al., 2009; Karydis et al., 2010; Athanasopoulou et al., 2010; Karydis et al., 2011a; Tsyro et al., 2011; Wang et al., 2012; Im, 2013; Trump et al., 2015). ~~Karydis et al. (2010; 2011a) showed that~~According to their findings, including marine and crustal species in models can substantially affect the phase partitioning ~~and size distribution~~ of nitrate aerosols. ~~Moreover, they showed that t~~Few of these studies have applied a hybrid approach for aerosol thermodynamics, which combines the dynamic calculation of mass transfer to coarse mode particles with an equilibrium approach for the fine mode particles, and found that mineral dust and sea salt can considerably affect the size distribution of aerosol nitrate (Athanasopoulou et al., 2008; Athanasopoulou et al., 2010; Karydis et al., 2010; Karydis et al., 2011a; Trump et al., 2015). The presence of mineral cations can also affect the aerosol ammonium concentrations due to thermodynamic interactions with the remainder ions in the aqueous phase (Karydis et al., 2010; 2011a). ~~Wang et al. (2012) found that the~~Furthermore, heterogeneous chemistry occurring on dust particles can also act as a source for sulfate (Wang et al., 2012). ~~Im (2013) identified the sea salt aerosol emissions as a significant source of aerosol nitrate in the Eastern Mediterranean coastal regions since they substantially increase the partitioning of nitric acid into the aerosol phase. Trump et al. (2015) applied a hybrid approach for aerosol thermodynamics over Europe, which combines the~~

~~dynamic calculation of mass transfer to coarse mode particles with an equilibrium approach for the fine mode particles, and they found that sea-salt emissions in areas with high nitric acid levels can reduce the fine nitrate aerosol concentrations.~~

The thermodynamic interactions of crustal elements with inorganic aerosol components can be very important on a global scale since mineral dust is a dominant compound in the atmosphere. Mineral dust accounts for more than 50% of the global aerosol load (Grini et al., 2005; Zender and Kwon, 2005) with Ca^{2+}, Mg^{2+}, K^+, and Na^+ in the form of mineral cations being the major chemically active components (Sposito, 1989). Dust particles largely originate from the subtropical deserts (Prospero et al., 2002) and can be transported over long distances and to high altitudes (Prospero et al., 2001; Kallos et al., 2007). The long-range transport of dust particles can influence the aerosol dynamics and atmospheric chemistry thousands of kilometers downwind of the source regions, while the chemical processing of the dust during transport can mobilize nutrients that are important for the marine biota (Solmon et al., 2009). Under favorable conditions dust particles from the Sahara desert can travel across the Mediterranean Sea toward Europe (Mitsakou et al., 2008; Querol et al., 2009; Bangert et al., 2011) or across the Atlantic Ocean toward the Caribbean (Chiapello et al., 2005; Kallos et al., 2006) and South America (Formenti et al., 2001), while dust from the Gobi and Taklimakan deserts often crosses the Pacific and can reach the west coast of the Americas (Fairlie et al., 2010; Wang et al., 2012; Karydis et al., 2011b). The dust particles can substantially influence air quality (Giannadaki et al., 2014). Therefore, an accurate representation of mineral dust emissions, transport, composition and chemistry is essential to minimize the nitrate aerosol related uncertainties in global chemistry-climate simulations.

However, most thermodynamic models used in global studies lack a realistic treatment of crustal species (Liao et al., 2003; Martin et al., 2003; Bauer et al., 2004; Koch et al., 2011; Leibensperger et al., 2012). Only a few global studies account for dust aerosol chemistry. Feng and Penner (2007) have included the heterogeneous reactions of HNO_3 with $CaCO_3$, $MgCO_3$, Na_2CO_3, and K_2CO_3 into a three dimensional aerosol and chemistry model to study the global distribution of nitrate and ammonium aerosol concentrations. Xu and Penner (2012) used the same model to explore the nitrate aerosol direct and indirect radiative forcing. Fairlie et al. (2010) have included the uptake of nitric acid on dust particles, limited by the dust alkalinity expressed as Ca^{2+}, on a global chemical transport model to study the impact of mineral dust on nitrate in transpacific Asian pollution plumes. Hauglustaine et al. (2014) applied the same uptake parametrization in a global model to simulate present and future nitrate aerosols and their climatic impact. However, these studies assumed a globally uniform chemical composition and size distribution for mineral dust particles and have not ~~focused on~~addressed the ~~actual~~ effect of mineral dust on nitrate aerosol formation and its tropospheric burden.

The present work ~~aims~~ is a first attempt to assess the effect of naturally emitted dust particles on global nitrate aerosol concentrations and size distributions. We have used ~~to improve the representation of nitrate aerosol formation and size distribution in~~ the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006) to simulate the nitrate aerosol formation and size distribution. ~~by including n~~Nitrate interactions with mineral dust~~,~~ have been taken into account by using the thermodynamic equilibrium model ISORROPIA II (Fountoukis and Nenes, 2007). ~~This study is a first attempt to assess the effect of naturally emitted dust particles on global nitrate aerosol concentrations and size distributions. For this purpose, a~~Dust emissions are calculated online by an advanced ~~online~~ dust emission scheme ~~is used~~ (Astitha et al., 2012)~~,~~ which accounts for the soil particle size distribution of different deserts worldwide. ~~Unique~~ C~~c~~hemical compositions of the emitted dust particles compiled from the literature are adopted for the main deserts to ~~enable~~ study the chemical interaction among crustal and inorganic species. Furthermore, the sensitivity of the results to the emitted dust aerosol load, the size distribution of the soil particles~~dust emission scheme based on the soil texture~~, the mineral dust chemical composition and the aerosol thermodynamic state is discussed.

2. Global Model Description

2.1 EMAC Model

We used the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model, which is a numerical chemistry and climate simulation system that includes sub-models describing ~~lower and middle~~ atmospheric processes from the troposphere to stratosphere and their interaction with oceans, land and human influences (Jöckel et al., 2006). It uses the Modular Earth Submodel System (MESSy2) (Jöckel et al., 2010) to link the different sub-models with ~~and~~ an ~~the~~ atmospheric dynamical core which is the 5th generation European Centre - Hamburg general circulation model

(ECHAM5) (Roeckner et al., 2006). 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 (Jöckel et al., 2006; Pozzer et al., 2006; de Meij et al., 2012; Pozzer et al., 2012). In this study, the spectral resolution of the EMAC model is T42L31, corresponding to a horizontal grid resolution of approximately 2.8^ox2.8^o and 31 vertical layers extending to 25 km altitude. EMAC is applied for 5 years covering the period 2004-2008 and the first year is used as spin-up.

The EMAC model calculates fields of gas phase species online through the Module Efficiently Calculating the Chemistry of the Atmosphere (MECCA) submodel (Sander et al., 2011). MECCA calculates the concentration of a range of gases, including aerosol precursor species such as SO_2, NH_3, NO_x, DMS, H_2SO_4 and DMSO. The concentrations of the major oxidant species (OH, H_2O_2, NO_3, and O_3) are also calculated online. The loss of gas phase species to the aerosol through heterogeneous reactions (e.g., N_2O_5 to form HNO_3) is treated using the MECCA_KHET submodel (Jöckel et al., 2010). The aqueous phase oxidation of SO_2 and the uptake of HNO_3 and NH_3 in cloud droplets are treated by the SCAV submodel (Tost et al., 2006; 2007).

Aerosol microphysics and gas/aerosol partitioning are calculated by the Global Modal-aerosol eXtension (GMXe) module (Pringle et al., 2010). The aerosol size distribution is described by 7 interacting lognormal modes (4 hydrophilic and 3 hydrophobic modes). ~~The modes cover the aerosol size spectrum (nucleation, Aitken, accumulation and coarse).~~ The aerosol composition within each mode is uniform with size (internally mixed), though can vary between modes (externally mixed). 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 removal of gas and aerosol species through wet deposition is calculated within the SCAV sub-model (Tost et al., 2006) while dry deposition is calculated within the DRYDEP submodel (Kerkweg et al., 2006) based on the big leaf approach.

The sedimentation of aerosols is calculated within the SEDI submodel (Kerkweg et al., 2006) using a first order trapezoid scheme.

2.2 Inorganic Aerosol Thermodynamics

The inorganic aerosol composition is computed with the ISORROPIA-II thermodynamic equilibrium model (Fountoukis and Nenes, 2007). ISORROPIA-II calculates the gas/liquid/solid equilibrium partitioning of the K^+-Ca^{2+}-Mg^{2+}-NH_4^+-Na^+-SO_4^{2-}-NO_3^--Cl^--H_2O aerosol system. Potassium, calcium, magnesium, and sodium are considered as chemically active components of mineral dust and are assumed to exist in the form of 14 mineral salts in the solid phase ($Ca(NO_3)_2$, $CaCl_2$, $CaSO_4$, $KHSO_4$, K_2SO_4, KNO_3, KCl, $MgSO_4$, $Mg(NO_3)_2$, $MgCl_2$, $NaHSO_4$, Na_2SO_4, $NaNO_3$, NaCl) and 4 ions in the aqueous phase (Ca^{2+}, K^+, Mg^{2+}, Na^+). ISORROPIA-II solves for the equilibrium state by considering the chemical potential of the species and minimizes the number of equations and iterations required by considering specific compositional "regimes". In ISORROPIA-II the aerosol can be in either a thermodynamically stable state (where salts precipitate once the aqueous phase becomes saturated) or in a metastable state (where the aerosol is composed only of a supersaturated aqueous phase). In this application we assume that aerosols can form solids (stable state) but we test the sensitivity of the results by using the metastable assumption as well. ~~Details about ISORROPIA-II can be found in Fountoukis and Nenes (2007).~~

The assumption of thermodynamic equilibrium is a good approximation for fine mode aerosols that 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 is 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.

2.3 Dust Emission Scheme

Dust emissions are calculated online by an advanced dust flux scheme developed by Astitha et al. (2012). The scheme uses the online meteorological fields from the EMAC model (temperature, pressure, relative humidity, soil moisture and the surface friction velocity) together with specific input fields for soil properties (i.e., the geographical location of the dust sources, the clay fraction of the soils, the rooting depth, and the monthly vegetation area index) and calculates the dust emission fluxes online. The dust particles are considered to be mobilized in the atmosphere when the wind friction velocity, a proxy of the surface drag properties, exceeds a threshold value. This threshold value depends on the soil size distribution and soil texture classification. The emission scheme uses an explicit geographical representation of the emitted soil particle size distribution based on soil characteristics in every grid cell (Astitha et al., 2012). The total annual average global emission flux of dust particles is 5684 Tg yr^{-1}. As a sensitivity study, an alternative dust emission scheme is used which utilizes a homogeneous global soil size distribution of dust particles and assumes that the emitted dust particles have the same size distribution in all grid cells based on D'Almeida (1987). In this case, the total annual average global emission flux of dust particles is 3660 Tg yr^{-1}. Details about the online dust production schemes used in this study can be found in Astitha et al. (2012).

Emissions of individual crustal species (Ca^{2+}, Mg^{2+}, K^{+}, Na^{+}) are estimated as a constant fraction of mineral dust emissions. This fraction is determined based on the geological information that exists for the different dust source regions of the planet and is applied online on the calculated mineral dust emissions based on the location of the grid cell. Table 1 lists the chemical composition of mineral dust used in this study for the main deserts of the world (Figure 1) based on the cited literature. As a sensitivity study, a spatially uniform mineral dust composition is also used by assuming a global emission ratio between crustal species and dust of 1.2%, 1.5%, 2.4%, and 0.9% for Na^{+}, K^{+}, Ca^{2+}, and Mg^{2+}, respectively, based on Sposito (1989).

2.4 Aerosol precursor and sea salt emissions

Fuel combustion and agriculture related emissions of NO_x, NH_3, and SO_2, which represent the gaseous precursors of the major inorganic components, are based on the high resolution (0.1°) global anthropogenic emission inventory applied at monthly intervals, EDGAR-CIRCE (Doering et al., 2009). The anthropogenic emissions are distributed vertically as described in Pozzer et al. (2009). The natural emissions of

NH_3 are based on the GEIA database (Bouwman et al., 1997) and include excreta from domestic animals, wild animals, synthetic nitrogen fertilizers, oceans, biomass burning, and emissions from soils under natural vegetation. NO_x produced by lightning is calculated online and distributed vertically based on the parameterization of Price and Rind (1992). The emissions of NO from soils are calculated online based on the algorithm of Yienger and Levy (1995). Eruptive and non-eruptive ~~Volcanic~~ volcanic degassing emissions of SO_2 are based on the AEROCOM data set (Dentener et al., 2006). The oceanic DMS emissions are calculated online by the AIRSEA submodel (Pozzer et al., 2006). The total global, annual average emissions of NO_x, NH_3, and SO_2 are 51 Tg yr^{-1}, 65 Tg yr^{-1}, and 65 Tg yr^{-1}, respectively. More details about the gas phase emissions used by EMAC can be found in Pozzer et al. (2012). Emissions of sea spray aerosols are based on the offline monthly emission data set of AEROCOM (Dentener et al., 2006) assuming a composition of 55% Cl^-, 30.6% Na^+, 7.7% SO_4^{2-}, 3.7% Mg^{2+}, 1.2% Ca^{2+}, 1.1% K^+ (Seinfeld and Pandis, 2006). The total global emission flux of sea spray aerosols is 5910 Tg yr^{-1}.

3 Model Evaluation

Tables 2, 3, and 4 include the comparison of model predictions of aerosol nitrate, sulfate, chloride, sodium, calcium, magnesium, and potassium concentrations with measurements from the European Monitoring and Evaluation Programme (EMEP; http://nilu.no/projects/ccc/emepdata.html), the Clean Air Status and Trends Network (CASTNET; http://java.epa.gov/castnet/timeframeselect.do), and the Acid Deposition Monitoring Network in East Asia (EANET; http://www.eanet.asia/product/index.html). The data selected for the evaluation is monthly averaged during the simulation period 2005-2008. The mean bias (MB), mean absolute gross error (MAGE), normalized mean bias (NMB), normalized mean error (NME), and the root mean square error (RMSE) were calculated to assess the model performance:

$$MAGE = \frac{1}{N}\sum_{i=1}^{N}\left|P_i - O_i\right| \qquad MB = \frac{1}{N}\sum_{i=1}^{N}\left(P_i - O_i\right)$$

$$NME = \frac{\sum_{i=1}^{N} |P_i - O_i|}{\sum_{i=1}^{N} O_i} \qquad NMB = \frac{\sum_{i=1}^{N} (P_i - O_i)}{\sum_{i=1}^{N} O_i}$$

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^{N} (P_i - O_i)^2 \right]^{\frac{1}{2}},$$

where P_i is the predicted value of the pollutant concentration, O_i is the observed value of the pollutant at the same monthly averaged time, and N is the total number of data points used for the comparison. NME and MAGE (in μg m^{-3}) provide an assessment of the overall discrepancy between model predictions and observations, while NMB and MB (in μg m^{-3}) are indicative of systematic errors. RMSE (in μg m^{-3}) incorporates both the variance of the prediction and its bias.

3.1 Mineral dust components

The model reproduces remarkably well the mineral dust cations (K^+, Mg^{2+}, Ca^{2+}) measured by the EMEP network (RMSE ~0.1 μg m^{-3}) indicating that their representation over the Sahara desert (e.g., mineral dust chemical composition and emission fluxes) proposed by this study is of the correct magnitude and is suitable to be used to estimate their effect on nitrate aerosol formation over Europe. Similarly, the model performs well in reproducing the mineral dust cations over North America (Table 3) indicating that the emissions from the deserts of the western USA suggested by this study are close to reality. The concentrations of K^+ and Mg^{+2} are reproduced well by the model (RMSE =0.05 μg m^{-3}) while Ca^{+2} is underestimated with NMB=-0.17. In contrast to Europe and North America, mineral cations are under-predicted by the model over Eastern Asia, especially calcium (NMB=-0.7), indicating that the Central Asian deserts could have a larger impact than assumed in this study.

3.2 Nitrate

EMAC systematically overpredicts nitrate concentrations compared to measurements from the EMEP network (MB=0.88 μg m^{-3}). This discrepancy is not the result of errors in the partitioning of the available nitric acid since EMAC significantly overestimates the total nitric acid concentrations (MB=2.29 μg m^{-3}). However, considering that routine nitrate filter-based measurements could be low biased due to the partial evaporation of nitrate from the teflon filters (Ames and Malm, 2001; Hering and Cass, 1999), notably at high temperatures in summer, some of this discrepancy can be attributed to the measurements as well. The model has the best overall performance statistically when compared against measurements from the CASTNET network, in comparison to the model performance over the EMEP and EANET networks. This is quite encouraging given that the CASTNET sites are located in rural areas and are not affected directly by urban sources which cannot be adequately captured by EMAC due to its coarse spatial resolution. Nitrate is unbiased when compared to the CASTNET network (MB=-0.04 μg m^{-3}) indicating that the model is successful in reproducing the relatively low nitrate levels over the continental U.S. (~0.8 μg m^{-3}). However, the NME is equal to 0.92 which indicates a high scatter. Considering that filter-based nitrate measurements are uncertain by roughly ±0.5 μg m^{-3} (Solomon et al., 2001; Karydis et al., 2007), this discrepancy at low nitrate concentrations could also be partially related to the measurements. Furthermore, there is little day-to-day variation of the emissions in the model and this simplification probably adds to the scatter as well. The model underpredicts all aerosol components over Eastern Asia, including nitrate, since it cannot capture the high concentrations observed over the urban centers of the Asian megacities (i.e., Beijing) due to its limited spatial resolution. Further, we apply emissions for 2005, which are probably low-biased for the following years considering the rapid growth of emissions in Asia. The underestimation of mineral cation emissions is responsible for the under-prediction of nitrate aerosol concentrations (MB=-0.69 μg m^{-3}) over Eastern Asia. Therefore, the impact of mineral dust on nitrate aerosol formation over Asia calculated by this study is probably underestimated as well.

3.3. Rest inorganic aerosol components

Over Europe, ammonium is slightly over-predicted (NMB=0.33) at least partly driven by the over-prediction of sulfate and nitrate concentrations (Table 2). On the other hand, sodium and chloride are slightly under-predicted by the model (MB=-0.26 μg m^{-3} and -0.31 μg m^{-3}, respectively) indicating a possible underestimation of sea salt emissions or too rapid removal during transport. Over North America, sulfate and ammonium are underestimated by the model; however, the model performance is better compared to other components (Table 2). Observed chloride concentrations are very low (0.05 µg m^{-3}). While the simulated concentrations are also low, they are systematically higher than the measurements with a MB of 0.32 µg m^{-3}. Over Eastern Asia, sulfate is significantly underpredicted (NMB=-0.67) since the observed concentrations are systematically high (i.e., 60 µg m^{-3} over Ha Noi during April 2007), not captured by the model. This results in an under-prediction of ammonium concentrations as well (NMB=-0.59) since ammonium is mostly sensitive to sulfate concentrations. Chloride concentrations are slightly under-predicted by the model (NMB=-0.21), however, their significant error (NME=1.03) indicates a high scatter.

4. Model Results

4.1 Mineral dust

The modeled global ~~inter-annual~~multi-year average surface concentration of mineral dust is 24 µg m^{-3} (Figure 2~~1~~a). High concentrations of mineral dust are calculated over the deserts (e.g., 1600 µg m^{-3} over the Bodele Depression, Sahara) and partly transported over very long distances (Figure 2~~1~~a). Dust particles originating from the Sahara desert can travel across the tropical Atlantic Ocean (10-90 µg m^{-3}) and across the Mediterranean affecting air quality in southern Europe (10-60 µg m^{-3}). The northwestern USA is mostly affected by dust originating from the Great Basin, Mojave, and Sonoran Deserts (60-440 µg m^{-3}). Dust concentrations can also be enhanced over the Pacific Ocean off the coast of California (2-15 µg m^{-3}). Dust from the Arabian (90-3000 µg m^{-3}), Thar (150-5000 µg m^{-3}), Taklimakan (250-9000 µg m^{-3}), and Gobi (70-1900 µg m^{-3}) deserts significantly affects air quality over the Middle East and Asia (250 µg m^{-3} on average). Mineral dust from the Thar and Arabian deserts affect the atmosphere over the Arabian Sea (15-100 µg m^{-3}). Over the Yellow Sea and North Pacific Ocean, dust concentrations are predicted to be 2-30 µg m^{-3} due to the long-range transport of dust from the Taklimakan and Gobi deserts. Dust-affected regions in the southern hemisphere are found in South America, e.g., from the Atacama (600-5000 µg m^{-3}) and Patagonian (250-2000 µg m^{-3}) deserts; Australia, e.g., from the Great Sandy and Simpson deserts (20-200 µg m^{-3}), and South Africa, e.g., from the Kalahari (100-700 µg m^{-3}) and Namibian deserts (100-2700 µg m^{-3}). Mineral dust from the Patagonian desert is efficiently transported over the South

Atlantic Ocean (15-150 μg m^{-3}) due to winds associated with the Antarctic circumpolar vortex that flow eastward around Antarctica. The dust emissions generated in S~~. ~~outh America are higher than what is stated in the literature and from satellite images (i.e., MODIS). As discussed in Astitha et al. (2012), the main reasons behind this over-prediction are the coarse model resolution in a region with pronounced topography, ~~while ~~which is a consequence of applying a consistent emission scheme throughout the globe without the use of regionally tuned emission fluxes.

4.1.1 Calcium

The global ~~inter-annual~~multi-year average surface concentration of calcium is 3.2 μg m^{-3} (Figure 2~~1~~b). The highest calcium concentrations are predicted over the Taklimakan Desert (50-1500 μg m^{-3}) where mineral dust emissions are extremely high and rich in calcium (12%). Dust particles originating from the Namibian Desert are also rich in calcium (12%), which results in high aerosol concentrations over the area (50-800 μg m^{-3}). Over the dust belt (e.g., Sahara, Thar, Gobi) the fraction of calcium to mineral aerosols is around 7% and the predicted aerosol concentration is 10-200 μg m^{-3}. In the rest of the world (e.g., the Americas and Australia) the fraction of calcium in mineral aerosols is less than 5% and the concentration is less than 60 μg m^{-3} with the lowest values over Australia (0.5-2 μg m^{-3}). In these areas, high calcium concentrations are calculated only over the Atacama Desert (20-200 μg m^{-3}) due to very high dust aerosol emissions.

4.1.2. Potassium

The global ~~inter-annual~~multi-year average surface concentration of potassium is 1.4 μg m^{-3} (Figure 2~~1~~c). The spatial distribution of potassium is similar to that of calcium due to their common origin; however, its magnitude is lower since the fraction of potassium in mineral dust is lower than that of calcium over all deserts (Table 1). The highest potassium concentrations are predicted over the Taklimakan (20-600 μg m^{-3}) and Namibian (20-350 μg m^{-3}) deserts where mineral dust emissions are highest and consist of 5% potassium. Over the Sahara and Thar deserts, where mineral dust is also rich in potassium (~3%), concentrations are predicted to be 2-140 μg m^{-3}. Over other deserts the fraction of potassium in mineral aerosols is low (0.1-

2%) and the concentration is less than 50 μg m^{-3} with the lowest values predicted over Australia (0.5 μg m^{-3}).

4.1.3. Magnesium

Magnesium is considered the least important chemically active constituent of mineral dust (Table 1). The global ~~inter-annual~~multi-year average surface concentration of magnesium is 1.7 μg m^{-3} (Figure 2~~1~~d). The highest magnesium concentrations are predicted over the Namib Desert (40-630 μg m^{-3}) where mineral dust is rich in magnesium (9%). High magnesium concentrations also occur over the Taklimakan desert (10-400 μg m^{-3}) due to the high dust emissions over the area. Over the Sahara, magnesium concentrations are 2-90 μg m^{-3}, while over other desert areas of the world, levels are lower (mostly below 60 μg m^{-3}) since its fraction in the dust aerosols is less than 2%. In contrast to calcium and potassium, ~~Magnesium~~ magnesium constitute a nontrivial ~~is also~~ part of sea spray emissions (3.7%)~~,~~ which results in 1-2 μg m^{-3} of magnesium over the Southern Oceans (South Atlantic, Pacific and Southern Oceans) and the North Atlantic and Pacific Oceans.

4.1.4. Sodium

The global ~~inter-annual~~multi-year average surface concentration of sodium is 5.4 μg m^{-3} (Figure 2~~1~~e). Sodium has high concentrations both over the deserts, following the spatial distribution of the rest chemically active dust components, and over the oceans, since it represents an important constituent of sea salt (30.6%). The highest sodium concentrations are predicted over the Atacama Desert (100-700 μg m^{-3}) due to high mineral dust fractions of sodium (7%). High sodium concentrations also occur over the Taklimakan (10-400 μg m^{-3}), Namibian (10-200 μg m^{-3}) and Thar (5-100 μg m^{-3}) deserts. Over the Oceans, sodium concentrations are 2-15 μg m^{-3} with the highest concentrations over the Southern Oceans.

4.2. Nitrate

The global ~~inter-annual~~multi-year average surface concentration of aerosol nitrate is 0.34 μg m^{-3}. The predicted total (gaseous nitric acid and aerosol) nitrate is 2-3 μg m^{-3} over the continents and can exceed 5 μg m^{-3} in the industrialized areas of Europe, central and eastern Asia, North America, as well as over biomass burning regions in the tropics (Figure 3~~2~~a). The highest values are found in the vicinity of Beijing in northeastern China (~10 μg m^{-3}). Total marine~~Marine total~~ nitrate concentrations are 1-2 μg m^{-3} on average nearly everywhere over the North Atlantic and Pacific Oceans, i.e., significantly enhanced compared to the oceans of the Southern Hemisphere. Fine aerosol nitrate is calculated to be higher in densely populated areas over Europe, China, and the Eastern USA (1-3 μg m^{-3}), mostly produced from local photochemistry, and decreases with distance from the urban source areas due to dilution and deposition, remaining at low levels in surrounding areas (lower than 0.5 μg m^{-3}) (Figures 3~~2~~b and 3c). Simulated coarse aerosol nitrate is found to be enhanced over Southern Europe, the Arabian Peninsula, Central and Eastern Asia, and Southwestern U.S. (1-4 μg m^{-3}~~-3~~), where HNO_3 from anthropogenic sources interacts with mineral dust from the surrounding deserts and thus largely condenses onto the coarse mode (Figures 3b and 3~~2~~c). Over these areas, where sulfuric acid is high, the bulk equilibrium assumption can result in the 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 leave more particle surface available for nitrate condensation. Coarse mode aerosol nitrate is also high over Central Africa where HNO_3 from biomass burning is adsorbed on the surface of coarse soil particles from the Sahara desert. 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,~~It is worth mentioning that~~ the assumption of thermodynamic equilibrium in the coarse mode may result in an overprediction of coarse aerosol nitrate~~, since the equilibrium timescale for large particles is typically larger than the timestep of the model (Meng and Seinfeld, 1996)~~. Assuming bulk equilibrium only for the fine aerosols and a dynamical approach for coarse particles could eliminate a possible bias (Capaldo et al., 2000;Karydis et al., 2010). 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 can kinetically condense within the time step of the model, in the equilibrium calculations.

4.3. ~~Sulfate~~Rest inorganic aerosol components

4.3.1. Sulfate

The global ~~inter-annual~~multi-year average surface concentration of aerosol sulfate is 1.8 μg m^{-3} (Figure 4~~3~~a). The highest aerosol sulfate concentrations are predicted over the industrialized areas of East Asia (3-10 μg m^{-3}), Europe (3-8 μg m^{-3}), India (2-6 μg m^{-3}), and the Eastern U.S. (2-5 μg m^{-3}), mostly in the fine mode. Sulfate concentrations can also exceed 4 μg m^{-3} over the Mediterranean as a result of transport of sulfur species from Europe. Concentrations over remote continental areas are 1-2 μg m^{-3} nearly everywhere in the Northern Hemisphere. Over the oceans, aerosol sulfate is mostly in the coarse mode, associated with sea spray emissions, leading to concentrations around 3 μg m^{-3}. The highest concentrations (4-5 μg m^{-3}) are calculated around the Arabian Peninsula (i.e., over the eastern Mediterranean and Persian Gulf), off the northeastern American and Asian coasts, and over the Northern Atlantic Ocean. Relatively high concentrations (3-4 μg m^{-3}) are also found over the Southern Oceans due to high DMS emissions. The lowest marine aerosol sulfate concentrations, less than 1 μg m^{-3}, occur over the remote tropical Pacific and Indian Oceans.

4.3.2. Ammonium

The global ~~inter-annual~~multi-year average surface concentration of aerosol ammonium is 1.8 μg m^{-3} (Figure 4~~3~~b). Ammonium calculations are very sensitive to the ammonia emissions and the calculated sulfate and nitrate concentrations. Therefore, ammonium follows the spatial distribution of sulfate and nitrate with high concentrations over East Asia (3-10 μg m^{-3}), Europe (3-8 μg m^{-3}), India (2-6 μg m^{-3}), and Eastern U.S. (2-5 μg m^{-3}), mostly in the form of ammonium sulfate and ammonium bisulfate and secondarily in the form of ammonium nitrate. Ammonium is also high over the biomass burning regions in the tropics, mostly in the form of ammonium nitrate (3-10 μg m^{-3}). Over the oceans, ammonium concentrations are negligible.

4.3.3. Chloride

The global ~~inter-annual~~multi-year average surface concentration of aerosol chloride is 7.8 μg m^{-3} (Figure 4~~3~~c). The highest concentrations are predicted over the

Southern Oceans and the Northern Atlantic Ocean (20-25 μg m^{3}) due to ~~strong~~ large sea spray emissions caused by the strong winds in the storm tracks associated with the synoptic~~large~~-scale vortices that circumvent the poles. Over the equatorial regions the chloride concentrations are 5-10 μg m^{-3}. Over the continents, chloride concentrations are high close to the coasts (2-7 μg m^{3}) and decrease rapidly with distance over land due to deposition processes.

~~5. Model Evaluation~~

~~Tables 2, 3, and 4 include the comparison of model predictions of fine aerosol nitrate, sulfate, chloride, sodium, calcium, magnesium, and potassium concentrations with measurements from the European Monitoring and Evaluation Programme (EMEP; http://nilu.no/projects/ccc/emepdata.html), the Interagency Monitoring of protected Visual Environments (IMPROVE; http://vista.cira.colostate.edu/improve/Data/IMPROVE/improve_data.htm), and the Acid Deposition Monitoring Network in East Asia (EANET; http://www.eanet.asia/product/index.html). The data selected for the evaluation is monthly averaged during the simulation period 2005-2008. The mean bias (MB), mean absolute gross error (MAGE), normalized mean bias (NMB), normalized mean error (NME), and the root mean square error (RMSE) were calculated to assess the model performance:~~

$$MAGE = \frac{1}{N}\sum_{i=1}^{N}\left|P_i - O_i\right| \qquad MB = \frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)$$

$$NME = \frac{\sum_{i=1}^{N}\left|P_i - O_i\right|}{\sum_{i=1}^{N}O_i} \qquad NMB = \frac{\sum_{i=1}^{N}(P_i - O_i)}{\sum_{i=1}^{N}O_i}$$

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)^2\right]^{\frac{1}{2}};$$

~~where P_i is the predicted value of the pollutant concentration, O_i is the observed value of the pollutant at the same monthly averaged time, and N is the total number of data points used for the comparison. NME and MAGE (in μg m^{-3}) provide an assessment of the overall discrepancy between model predictions and observations, while NMB~~

~~and MB (in µg m^{-3}) are indicative of systematic errors. RMSE (in µg m^{-3}) incorporates both the variance of the prediction and its bias.~~

~~5.3. Europe~~

~~EMAC systematically overpredicts nitrate concentrations compared to measurements from the EMEP network (MB=0.88 µg m^{-3}). This discrepancy is not the result of errors in the partitioning of the available nitric acid. Sulfate concentrations are actually overpredicted (MB=1.36 µg m^{-3}) and most of the cations are captured very well by the model (Table 2). Therefore, it is possible that EMAC overestimates the total nitric acid concentrations. However, considering that routine nitrate filter-based measurements could be low biased due to the partial evaporation of nitrate from the teflon filters (Ames and Malm, 2001; Hering and Cass, 1999), notably at high temperatures in summer, some of this discrepancy can be attributed to the measurements as well. Ammonium is slightly over-predicted by EMAC (NMB=0.33) at least partly driven by the over-prediction of sulfate and nitrate concentrations. On the other hand, sodium and chloride are slightly under-predicted by the model (MB=-0.26 µg m^{-3} and -0.31 µg m^{-3}, respectively) indicating a possible underestimation of sea salt emissions or too rapid removal during transport. The other mineral dust cations (K^+, Mg^{2+}, Ca^{2+}) are reproduced remarkably well by the model (RMSE ~0.1 µg m^{-3}) indicating that their representation over the Sahara desert proposed by this study is of the correct magnitude and is suitable to be used to estimate their effect on nitrate aerosol formation over Europe.~~

~~5.4. North America~~

~~The model has the best overall performance statistically when compared against measurements from the IMPROVE network. This is quite encouraging given that the IMPROVE sites are located in rural areas and are not affected directly by urban sources which cannot be adequately captured by EMAC due to its coarse spatial resolution. Furthermore, the high number of observational sites and the high frequency of measurements resulted in a data set of approximately 8,000 data points for each of the aerosol components. This makes the statistical analysis more reliable compared to the networks in Europe (EMEP) and Asia (EANET) which have approximately 1,000 data points including measurements from urban areas. Nitrate is unbiased when compared to the IMPROVE network (MB=-0.07 µg m^{-3}) indicating~~

~~that the model is successful in reproducing the relatively low nitrate levels over the continental U.S. (~0.5 μg m^{-3}). However, the NME is equal to 0.94 which indicates a high scatter. Considering that filter-based nitrate measurements are uncertain by roughly ±0.5 μg m^{-3} (Solomon et al., 2001; Karydis et al., 2007), this discrepancy at low nitrate concentrations could also be partially related to the measurements. Furthermore, there is little day-to-day variation of the emissions in the model and this simplification probably adds to the scatter as well. The performance of the model regarding sulfate is very good (NME=0.37). However, the model cannot capture the high sulfate concentrations from specific volcanic activities (i.e., 12 μg m^{-3} over Hawaii during February 2008) since EMAC uses background volcanic emissions (outgassing), which are identical for each year. Nevertheless, these points are rather limited in number and the overall bias of the model is low (NMB=-0.17). Ammonium is not systematically measured by the IMPROVE stations and the available observations are very few (only 62 data points). Observed chloride concentrations are very low (0.08 μg m^{-3}). While the simulated concentrations are also low, they are systematically higher than the measurements with a MB of 0.29 μg m^{-3}. Similar to Europe, the model performs well in reproducing the mineral dust cations over the USA (RMSE ~0.1 μg m^{-3}) indicating that their emissions from the deserts of the western USA suggested by this study are close to reality.~~

~~5.5. East Asia~~

~~The model underpredicts all aerosol components over Eastern Asia since it cannot capture the high concentrations observed over the urban centers of the Asian megacities (i.e., Beijing) due to its limited spatial resolution. Further, we apply emissions for 2005, which are probably low-biased for the following years considering the rapid growth of emissions in Asia. Sulfate is significantly underpredicted (NMB=-0.67) since the observed concentrations are systematically high (i.e., 60 μg m^{-3} over Ha Noi during April 2007), not captured by the model. This results in an under-prediction of ammonium concentrations as well (NMB=-0.59) since ammonium is mostly sensitive to sulfate concentrations. Chloride concentrations are slightly under-predicted by the model (NMB=-0.21), however, their significant error (NME=1.03) indicates a high scatter. In contrast to Europe and USA, mineral cations are under-predicted by the model over Eastern Asia, especially calcium (NMB=-0.7), indicating that the Central Asian deserts could have a larger impact than~~

~~assumed in this study. The underestimation of mineral cation emissions is probably at least partially responsible for the under-prediction of nitrate aerosol concentrations (MB=-0.69 μg m^{-3}) over Eastern Asia. Therefore, the impact of mineral dust on nitrate aerosol formation over Asia estimated by this study is probably a lower limit.~~

~~6.~~5. Mineral Dust Effect on Inorganic Aerosol

To estimate the effects of mineral dust on the inorganic aerosol concentration, on the phase partitioning of nitrate,~~ and on the nitrate aerosol concentration~~ and on the size distribution of nitrate aerosols, a sensitivity run was conducted~~ in which the presence of the reactive dust components has been ignored by~~ switching off the dust-aerosol chemistry. Therefore, in this sensitivity simulation mineral dust is considered to be a bulk species without chemical identity, and cations (Ca^{2+}, Mg^{2+}, K^+, Na^+) exist only as part of the sea salt aerosols. This is a dust configuration that is common in atmospheric chemistry transport and climate calculations.

~~6.3. Effects on phase partitioning of nitrate~~

~~Figure 4 shows the fraction of total nitrate occurring in the aerosol phase (~~ $NO_{3\ [Aerosol]}^{-} / \left(NO_{3\ [Aerosol]}^{-} + NO_{3\ [Gas]}^{-} \right)$ ~~) calculated by the base case and the sensitivity simulations. In areas where the dust concentrations are high (over the deserts), nitric acid is associated with non-volatile mineral cations (~~Na^+, Ca^{2+}, K^+, Mg^{2+}~~) forming salts in order to maintain the charge balance in the aerosol phase. The fraction of nitrate in the aerosol phase varies between 10% over the Great Basin desert to 90% over the Gobi desert where mineral dust is associated with nitric acid originating from the anthropogenic sources of Eastern Asia. Over Africa, the calculated nitrate aerosol fraction is 20%-60% with the highest values predicted over the equatorial region, which is affected by high mineral dust concentrations from the Sahara and enhanced nitric acid concentrations from biomass burning in the Congo Basin. In the sensitivity simulation where dust reactive components are ignored, nitric acid largely remains in the gas phase in areas close to deserts.~~

5.1Effects on inorganic aerosol concentrations

5.1.1 Effects on nitrate aerosols

The absolute and fractional changes of aerosol nitrate concentration at the surface between the base case and the sensitivity simulation are depicted in Figure 5. Positive changes correspond to higher concentrations in the base case~~ simulation~~. The predicted aerosol nitrate is higher in the base case simulation (up to 3 μg m^{-3}) due to the formation of salts with mineral components ($NaNO_3$, $Ca(NO_3)_2$, KNO_3, $Mg(NO_3)_2$). This does not take place in the sensitivity simulation where mineral dust is assumed to be chemically inert~~reactive components are ignored~~, and nitric acid remains in the gas phase. The predicted fractional change of nitrate aerosol concentration due to the interaction with mineral dust cations is up to 100% over the main deserts with the highest values calculated over the Saharan, Arabian, and Indian deserts. The relatively lowest fractional changes are calculated over the deserts of the Southern Hemisphere (i.e., Patagonia, Australia). The contribution of mineral dust to aerosol nitrate is not only important over areas with high dust concentrations but also downwind of the sources. For instance, across southern Europe the aerosol nitrate concentration increases due to the dust aerosol chemistry treatment by 0.5 μg m^{-3}, over western and eastern USA by 2 μg m^{-3} and 0.5 μg m^{-3}, respectively, over eastern China and northern India by 0.5 μg m^{-3}, and over Central Africa by 2 μg m^{-3}. Overall, the total predicted domain average nitrate aerosol concentration at the surface increases by 36% ~~after ~~by considering the interactions of nitrate with mineral dust cations.

The tropospheric burdens of ~~the main inorganic~~nitrate aerosols calculated in the base case and the sensitivity simulations are listed in Table 5. The nitrate aerosol tropospheric burden increases substantially by 0.2 Tg, i.e., 44%, by considering the dust aerosol chemistry. Moreover, the tropospheric burden of total nitrate (gaseous HNO_3 and aerosol nitrate) is 0.07 Tg (3%) lower in the base case simulation even though the NO_x emissions remain unchanged in the sensitivity test. This difference is due to the more efficient removal of total nitrate since the base case predicts a higher fraction of total nitrate in the aerosol phase compared to the sensitivity simulation. Nitrate aerosols are removed more efficiently through both dry and wet deposition compared to the gas phase HNO_3, especially the nitrate in coarse mode particles that are additionally removed by sedimentation.

5.1.2. Effects on the rest inorganic aerosol components

The tropospheric burdens of the main inorganic aerosols calculated in the base case and the sensitivity simulations are listed in Table 5. Chloride anions are ~~also~~ associated with the non-volatile mineral cations, which results in an increase of the aerosol chloride tropospheric burden by 0.3 Tg (9%). The tropospheric burden of ammonium decreases by 0.12 Tg (41%) due to dust aerosol chemistry even though it is not associated directly with the alkaline mineral components. This decrease can be attributed to the reduction of available nitric acid in the atmosphere due to the presence of the mineral cations, which leads to a decrease of ammonium nitrate production. Sulfate aerosol increases by 0.13 Tg (7%) by taking into account the mineral dust components. Sulfate is a non-volatile aerosol compound and exists in the particulate phase even in the form of H_2SO_4 and therefore its phase partition is not affected by the presence of cations. However, SO_4^{2-} can be formed heterogeneously in fogs and clouds via the dissolution of gaseous SO_2 and its oxidation by H_2O_2 or O_3. The reaction of the dissolved SO_2 with O_3 can be very important at pH values greater than about 5~~.35~~ (Seinfeld and Pandis, 2006) and therefore, the in-cloud oxidation rate of SO_2 can increase substantially in the presence of alkaline species such as the mineral cations that increase the pH.

5.2 Effects on phase partitioning of nitrate

Figure 6 shows the fraction of total nitrate occurring in the aerosol phase ($NO_3^-{}_{[Aerosol]} \big/ \left(NO_3^-{}_{[Aerosol]} + NO_3^-{}_{[Gas]} \right)$) calculated by the base case and the sensitivity simulations. In areas where the dust concentrations are high (over the deserts), nitric acid is associated with the non-volatile mineral cations (Na^+, Ca^{2+}, K^+, Mg^{2+}) forming salts in order to maintain the charge balance in the aerosol phase. The fraction of nitrate in the aerosol phase varies between 10% over the Great Basin desert to 90% over the Gobi desert where mineral dust is associated with nitric acid originating from the anthropogenic sources of Eastern Asia. Over Africa, the calculated nitrate aerosol fraction is 20%-60% with the highest values predicted over the equatorial region, which is affected by high mineral dust concentrations from the Sahara and enhanced nitric acid concentrations from biomass burning in the Congo Basin. In the sensitivity simulation where dust reactive components are ignored, nitric acid largely remains in the gas phase in areas close to deserts.

~~5.1~~5.3 Effects on nitrate aerosol size distribution

The fraction of aerosol nitrate in the coarse mode increases in the base case simulation since most of the mineral cations occur in the coarse mode. The model predicts that about 50% of the global mean total aerosol nitrate is in the coarse mode. In the sensitivity simulation in which mineral dust is assumed to be chemically inert~~the presence of reactive dust components is ignored~~, the corresponding fraction of coarse mode nitrate to total aerosol nitrate is 44%. Over the deserts, the fraction of nitrate in the coarse mode is nearly 100% and declines with distance from the dust source regions. Since the model assumes that equilibrium is established separately for each mode, the presence of mineral cations in the coarse mode traps nitric acid vapor thus lowering the nitric acid concentration in the gas phase. The fine aerosol then loses mass as evaporation is required to maintain equilibrium with the gas phase. As a result, the predicted fine aerosol nitrate may occasionally decrease in the presence of mineral dust. However, over areas where nitric acid is not the limiting reactant, nitrate increases in the fine mode, since a fraction of mineral dust exists in the fine mode as well. Overall, the domain average nitrate aerosol concentration at the surface increases by 21% in the fine mode and 53% in the coarse mode by considering the interactions of nitrate with mineral dust cations.

6 Sensitivity Tests

We have conducted four additional sensitivity simulations to investigate if the nitrate aerosol formation depends strongly on i) the dust emission parameterization scheme, ii) the chemical composition of the emitted dust aerosols, iii) the strength of the dust aerosol emissions, and iv) the aerosol state assumption. Figure ~~6~~ 7 depicts the inter-annual absolute change of aerosol nitrate concentrations compared to the base case for each of the sensitivity simulations. ~~Positive change corresponds to higher concentrations in the base case simulation.~~ A positive change corresponds to a decrease of the concentrations in the sensitivity simulations. The tropospheric burdens of the main inorganic aerosols from each of the sensitivity simulations are listed in Table 5.

6.1 Sensitivity to the dust emission parameterization scheme

The first sensitivity test utilizes a homogeneous global soil size distribution of dust particles, in contrast to the base case simulation that uses an explicit geographical representation. Another difference between the two simulations is the emitted particle size distribution at the source; in the sensitivity case the D'Almeida (1987) “background” source modes are imposed uniformly in all grid cells whereas the base case explicitly accounts for the soil characteristics in every grid cell. This influences the calculated threshold friction velocity, which triggers the dust mobilization and hence changes the dust aerosol emission fluxes. Consequently, the tropospheric burdens of mineral components calculated by the sensitivity case simulation differ substantially from the base case simulation (Table 5). 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 (Astitha et al., 2012). The reduced emissions of mineral components in the sensitivity case simulation result in a decrease of the tropospheric nitrate burden by 9%. The largest absolute decrease is calculated over northeastern China (0.7 μg m^{-3}, 15%), which is affected by mineral dust emitted from the Central Asian deserts. The highest fractional decrease is calculated over the eastern Amazon Basin (0.4 μg m^{-3} or 40%) affected by dust from the Atacama Desert. The reduction of nitrate in the sensitivity simulation is also important over the Western U.S. (0.4 μg m^{-3}, 30%). On the other hand, over the Sahara the sensitivity case simulation predicts higher emissions of mineral components, which result in an increase of nitrate aerosol concentrations over the Congo Basin by 0.1 μg m^{-3} (10%).

6.2 Sensitivity to the emitted dust aerosol composition

The second sensitivity test assumes a globally uniform chemical composition of mineral dust in contrast to the base case simulation where the mineral dust composition depends~~varies based~~ on the soil characteristics of each desert. While the emitted total mineral dust aerosols remain the same between the two simulations, the different assumptions on the mineral dust chemical composition result in significant changes ~~on~~ in the calculated tropospheric burden of the individual mineral dust components (Table 5). The fraction of the individual mineral components to total mineral dust assumed in the sensitivity simulation is lower in dust from most of the deserts compared to the base case (Table 1), which results in reduced emissions. This substantially affects not only the tropospheric burden of the mineral components (Na^+,

Ca^{+2}, K^{+}, and Mg^{+2}, are reduced by 17%, 40%, 37%, and 48%, respectively), but also the calculated tropospheric burden of nitrate aerosol, which decreases by 16% in the sensitivity simulation. The largest absolute decrease is calculated over northeastern China (1 μg m^{-3} or 20%). The highest fractional decrease is calculated over the Congo Basin (0.6 μg m^{-3}, 60%). Over the western USA nitrate decreases by 0.5 μg m^{-3} or 35%). On the other hand, nitrate aerosol concentrations are predicted to increase close to the Atacama Desert (0.1 μg m^{-3}, 30%).

6.3 Sensitivity to the emitted dust aerosol load

The third sensitivity test assumes 50% lower emissions of mineral dust aerosol compared to the base case simulation, and is used to estimate the corresponding effect on the nitrate aerosol formation. Despite the drastic decrease of the atmospheric dust load (43%), the tropospheric nitrate burden decreases by only 9% (Table 5). This is not unexpected since the thermodynamic interactions between nitrate and mineral components are mostly important over the deserts where nitric acid is the limiting reactant rather than the mineral dust. However, over areas that are located close to the main deserts and are at the same time rich in nitric acid, the impact of the dust emission reduction on the nitrate formation is substantial. For instance, nitrate aerosol concentrations decrease significantly over northeastern China (0.7 μg m^{-3}, 15%), the Congo Basin (0.5 μg m^{-3}, 40%), southern Europe (0.3 μg m^{-3}, 15%) and the USA (0.4 μg m^{-3}, 30%). The largest absolute decrease is calculated over northern India (0.7 μg m^{-3}, 25%). The highest fractional decrease is calculated close to the Atacama Desert (0.15 μg m^{-3}, 55%).

6.4 Sensitivity to the aerosol state assumption

The final sensitivity test assumes that the aerosol solution is aqueous even at very low relative humidity (metastable assumption), and it is used to estimate the impact of the aerosol phase state on the nitrate aerosol formation. The calculated tropospheric burden of nitrate aerosol decreases by only 2% in the sensitivity simulation. Ansari and Pandis (2000) suggested that the stable state (assumed in our base case simulation) results in higher concentrations of aerosol nitrate when the RH is very low (< 35%) and/or sulfate to nitrate molar ratios are low (< 0.25). This results in a decrease of the calculated nitrate aerosol concentrations close to deserts in the sensitivity simulation. The largest absolute decrease (0.3 μg m^{-3}, 20%) is calculated around the Central Asian deserts (~~0.3 μg m^{-3}, 20%~~sulfate/nitrate=0.3; RH=20%). The highest fractional decrease (0.2 μg m^{-3}, 60%) is calculated close to the Atacama Desert (sulfate/nitrate=0.4; RH=28%~~0.2 μg m^{-3}, 60%~~). On the other hand, over areas where the sulfate to nitrate molar ratio is high (i.e., over eastern China, northeastern U.S.), or when the relative humidity is very high (i.e., over the Congo Basin), the metastable assumption results in higher nitrate aerosol concentrations. The largest absolute increase (0.15 μg m^{-3}, 5%) is calculated over ~~north~~eastern China (sulfate/nitrate=0.9; RH=86%~~0.15 μg m^{-3}, 5%~~) and the highest fractional increase (0.1 μg m^{-3}, 10%) over the Congo Basin (sulfate/nitrate=0.2; RH=92%~~0.1 μg m^{-3}, 10%~~).

7 Summary and ~~Conclusions~~conclusions

This study assesses the effect of mineral dust particles on nitrate aerosol formation by using the thermodynamic equilibrium model ISORROPIA-II that takes the thermodynamics of the $K^+\text{-}Ca^{2+}\text{-}Mg^{2+}\text{-}NH_4^+\text{-}Na^+\text{-}SO_4^{2-}\text{-}NO_3^-\text{-}Cl^-\text{-}H_2O$ components into account. The fine aerosol nitrate concentration is predicted to be higher over highly populated and industrialized areas (up to 3 μg m^{-3}), while coarse aerosol nitrate is found to be higher over the deserts (up to 4 μg m^{-3}).

The contribution of mineral dust to nitrate aerosol concentrations is significant in areas with high dust concentrations (near deserts) with impacts that can extend across southern Europe, western USA and northeastern China. Over these areas, nitric acid is associated with non-volatile mineral cations (Na^+, Ca^{2+}, K^+, Mg^{2+}) forming salts to maintain the charge balance in the aerosol phase. This is not reflected in the sensitivity simulation where dust reactive components are ignored and nitric acid remains solely in the gas phase in areas close to deserts. As a consequence, 36% higher global average nitrate aerosol concentrations are produced at the surface~~ by 36%~~ in the base case simulation, while the coarse and fine mode nitrate concentrations are higher by 53% and 21%, respectively. The tropospheric burden of nitrate aerosol increases by 44% when considering dust aerosol chemistry. Given that all results from this study are reported as multi-annual averages, this contribution can be even more important during strong dust ~~storm~~ episodes.

~~The tropospheric burden of nitrate aerosol increases by 44% when considering dust aerosol chemistry.~~ Other inorganic aerosol components are affected by the presence of the reactive dust components as well. Chloride is directly associated with the mineral cations and its tropospheric burden increases by 9%. The tropospheric burden of ammonium decreases by 41% due to the reduction of available nitric acid in the gas phase. The tropospheric burden of sulfate increases by 7% as the pH dependent in-cloud oxidation of SO_2 by O_3 increases due to the presence of alkaline mineral dust components.

Four additional simulation tests have been conducted to investigate the sensitivity of the results to the mineral dust emission parameterization scheme, the chemical composition of the emitted dust, the emitted dust aerosol load and the aerosol state assumption. These simulations indicate that the calculated nitrate ~~aerosol formation~~tropospheric burden is mostly sensitive to the chemical composition of mineral dust. By assuming a global uniform chemical composition of mineral dust, we ~~found~~ find a ~~change~~ reduction of 16% in the calculated tropospheric burden of nitrate aerosol. ~~The largest differences are predicted over northeastern China (1 μg m^{-3}, 20%) and the Congo Basin (0.6 μg m^{-3}, 60%).~~ The results are moderately sensitive to the mineral dust aerosol load and the mineral dust emission scheme as the dust itself is often not the limiting factor (in both cases a 9% change in the nitrate aerosol tropospheric burden is calculated). The aerosol state assumption has a marginal effect on the calculated nitrate aerosol tropospheric burden (2% change); however, it can be important over deserts (stable state assumption predicts higher nitrate concentrations) and sulfate rich areas (metastable state assumption predicts higher nitrate concentrations).

The relative importance of the examined parameters to the nitrate aerosol formation is not spatially uniform. The calculated nitrate aerosol concentrations are mostly sensitive to the mineral dust chemical composition over areas close to deserts that are rich in nitric acid (i.e., nitrate is reduced by 60% over the Congo Basin, 35% over the western USA, and 20% over the northeastern China). The aerosol state assumption is the most influential parameter in simulations of the nitrate aerosol formation over deserts at very low relative humidity (i.e., nitrate is reduced by 60% over the Atacama Desert where RH=28% and 20% over the Central Asian deserts where RH=20%). Mineral dust emissions are the dominant factor for simulations of the nitrate aerosol formation over southern Europe (nitrate is reduced by 15%) and northern India (nitrate is reduced by 25%) while the size distribution of mineral dust is mostly important over eastern Amazon Basin (nitrate is reduced by 40%).

By~~We~~ ~~used~~ using an explicit geographical representation of the emitted soil particle size distribution and chemical composition based on soil characteristics of each desert we~~to~~ have ~~quantify~~ quantified the impact of mineral dust on nitrate aerosol formation due to thermodynamical interactions (assuming equilibrium). Despite the sensitivities and associated uncertainties, we conclude that mineral dust aerosol chemistry is important for nitrate aerosol formation. Neglecting the thermodynamic interactions of nitrate with mineral cations may introduce significant biases in the global distribution of nitrate as well as other aerosol components, especially for coarse mode aerosols. Given that the coating of dust by hygroscopic salts affects its efficiency to grow by water uptake and act as cloud condensation nuclei, the ability to capture the complex interactions of mineral dust with the inorganic aerosol components is of prime importance for global and regional air quality and climate models. The role of mineral dust on the direct aerosol effect as well as its effect on the cloud condensation nuclei formation will be investigated in a ~~planned~~ future study.

Acknowledgements

The research leading to these results has received funding from the European Research Council under the European Union's Seventh Framework Programme (FP7/2007-2013) / ERC grant agreement n° 226144. V.A. Karydis acknowledges support from a FP7 Marie Curie Career Integration Grant (project reference 618349). A.P. Tsimpidi acknowledges support from a DFG individual grand programme (project reference TS 335/2-1).

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Table 1. Chemical composition of mineral dust

	Crustal Species					
Desert	Na^+	K^+	Ca^{2+}	Mg^{2+}	Other	***Reference***
Great Basin	0.064	0.023	0.053	0.018	0.842	(Fantle et al., 2012)
Mojave	0.015	0.027	0.059	0.019	0.880	(Reynolds et al., 2006)
Sonoran	0.025	0.012	0.037	0.006	0.920	(Kasper-Zubillaga and Zolezzi-Ruiz, 2007)
Patagonia	0.012	0.015	0.021	0.013	0.939	(Gaiero et al., 2007)
Monte	0.023	0.018	0.025	0.009	0.925	(Tripaldi et al., 2010)
Atacama	0.069	0.007	0.018	0.005	0.901	(Michalski et al., 2004)
Kalahari/ Namibia	0.030	0.050	0.120	0.090	0.710	(Resane et al., 2004)
Sahara	0.011	0.035	0.075	0.030	0.849	(Formenti et al., 2008)
Saudi Arabia	0.010	0.004	0.034	0.006	0.946	(Dada et al., 2013)
Thar/Lut	0.022	0.033	0.082	0.022	0.841	(Yadav and Rajamani, 2004)
Taklimakan	0.012	0.030	0.120	0.028	0.810	(Zhang et al., 2003)
Gobi	0.012	0.021	0.077	0.017	0.873	(Zhang et al., 2003)
Great Sandy/ Simpson	0.028	0.001	0.005	0.003	0.963	(Radhi et al., 2011)
Other	0.012	0.015	0.024	0.009	0.940	(Sposito, 1989)

	~~***Crustal Species***~~					
~~***Desert***~~	~~Na^+~~	~~K^+~~	~~Ca^{2+}~~	~~Mg^{2+}~~	~~Dust~~	~~***Reference***~~
~~Great Basin~~	~~0.064~~	~~0.023~~	~~0.053~~	~~0.018~~	~~0.842~~	~~(Fantle et al., 2012)~~
~~Mojave~~	~~0.015~~	~~0.027~~	~~0.059~~	~~0.019~~	~~0.880~~	~~(Reynolds et al., 2006)~~
~~Sonoran~~	~~0.025~~	~~0.012~~	~~0.037~~	~~0.006~~	~~0.920~~	~~(Kasper-Zubillaga and Zolezzi-Ruiz, 2007)~~
~~Patagonia~~	~~0.012~~	~~0.015~~	~~0.021~~	~~0.013~~	~~0.939~~	~~(Gaiero et al., 2007)~~
~~Monte~~	~~0.023~~	~~0.018~~	~~0.025~~	~~0.009~~	~~0.925~~	~~(Tripaldi et al., 2010)~~
~~Atacama~~	~~0.069~~	~~0.007~~	~~0.018~~	~~0.005~~	~~0.901~~	~~(Michalski et al., 2004)~~
~~Kalahari/ Namib~~	~~0.030~~	~~0.050~~	~~0.120~~	~~0.090~~	~~0.710~~	~~(Resane et al., 2004)~~
~~Sahara~~	~~0.01~~	~~0.035~~	~~0.075~~	~~0.030~~	~~0.849~~	~~(Formenti et al., 2008)~~

	~~1~~					
~~Saudi Arabia~~	~~0.010~~	~~0.004~~	~~0.034~~	~~0.006~~	~~0.946~~	~~(Dada et al., 2013)~~
~~Thar/Lut~~	~~0.022~~	~~0.033~~	~~0.082~~	~~0.022~~	~~0.841~~	~~(Yadav and Rajamani, 2004)~~
~~Taklimakan~~	~~0.012~~	~~0.030~~	~~0.120~~	~~0.028~~	~~0.810~~	~~(Zhang et al., 2003)~~
~~Gobi~~	~~0.012~~	~~0.021~~	~~0.077~~	~~0.017~~	~~0.873~~	~~(Zhang et al., 2003)~~
~~Great Sandy/ Simpson~~	~~0.028~~	~~0.001~~	~~0.005~~	~~0.003~~	~~0.963~~	~~(Radhi et al., 2011)~~
~~Other~~	~~0.012~~	~~0.015~~	~~0.024~~	~~0.009~~	~~0.940~~	~~(Sposito, 1989)~~

Table 2. Statistical evaluation of EMAC simulated aerosol concentrations against monthly average observations from Europe during 2005–2008.

EMEP Network

Metric	NO_3^-	Na^+	Ca^{2+}	K^+	Mg^{2+}	NH_4^+	Cl^-	SO_4^{2-}
Observed (μg m^{-3})	0.36	0.91	0.13	0.11	0.09	0.72	1.31	0.64
Calculated (μg m^{-3})	1.24	0.65	0.12	0.06	0.1	1.04	1	2
MAGE (μg m^{-3})	0.91	0.45	0.07	0.06	0.06	0.43	0.59	1.34
MB (μg m^{-3})	0.88	-0.26	-0.01	-0.05	0.01	0.32	-0.31	1.33
NME	1.98	0.49	0.57	0.55	0.66	0.6	0.45	1.77
NMB	1.83	-0.01	-0.01	-0.35	0.46	0.33	-0.24	1.75
RMSE (μg m^{-3})	0.96	1.72	0.13	0.11	0.08	0.79	0.67	1.36
number of comparisons	1455	1121	1479	1400	1266	1450	423	2792

Table 3. Statistical evaluation of EMAC simulated aerosol concentrations against monthly average observations from North America during 2005–2008.

CASTNET Network

Metric	NO_3^-	Na^+	Ca^{2+}	K^+	Mg^{2+}	NH_4^+	Cl^-	SO_4^{2-}
Observed (μg m^{-3})	0.86	0.09	0.29	0.06	0.04	1.04	0.05	2.81
Calculated (μg m^{-3})	0.82	0.27	0.12	0.05	0.06	0.85	0.37	2.11
MAGE (μg m^{-3})	0.8	0.19	0.22	0.03	0.04	0.45	0.34	1.05
MB (μg m^{-3})	-0.04	0.18	-0.17	-0.01	0.02	-0.19	0.32	-0.71
NME	0.92	2.09	0.75	0.53	0.92	0.43	6.36	0.37
NMB	-0.05	1.94	-0.58	-0.13	0.4	-0.18	6.13	-0.25

RMSE (µg m^{-3})	1.19	0.24	0.38	0.05	0.05	0.06	0.5	1.5
number of comparisons	1523	1523	1523	1522	1523	1523	1523	1523

~~**IMPROVE Network**~~

~~**Metric**~~	~~NO_3^-~~	~~Na^+~~	~~Ca^{2+}~~	~~K^+~~	~~Mg^{2+}~~	~~NH_4^+~~	~~Cl^-~~	~~SO_4^{2-}~~
~~**Observed** (µg m^{-3})~~	~~0.51~~	~~0.12~~	~~0.05~~	~~0.04~~	~~0.03~~	~~1.11~~	~~0.08~~	~~1.49~~
~~**Calculated** (µg m^{-3})~~	~~0.44~~	~~0.32~~	~~0.13~~	~~0.06~~	~~0.07~~	~~0.86~~	~~0.37~~	~~1.28~~
~~**MAGE** (µg m^{-3})~~	~~0.48~~	~~0.22~~	~~0.1~~	~~0.04~~	~~0.05~~	~~0.51~~	~~0.31~~	~~0.55~~
~~**MB** (µg m^{-3})~~	~~-0.07~~	~~0.20~~	~~0.08~~	~~0.02~~	~~0.04~~	~~-0.25~~	~~0.29~~	~~-0.21~~
~~**NME**~~	~~0.94~~	~~1.88~~	~~2~~	~~0.82~~	~~1.84~~	~~0.46~~	~~4.08~~	~~0.37~~
~~**NMB**~~	~~-0.18~~	~~1.62~~	~~1.63~~	~~0.33~~	~~1.57~~	~~-0.23~~	~~3.74~~	~~-0.17~~
~~**RMSE** (µg m^{-3})~~	~~0.91~~	~~0.27~~	~~0.11~~	~~0.06~~	~~0.05~~	~~0.58~~	~~0.49~~	~~0.94~~
~~**number of comparisons**~~	~~8108~~	~~8073~~	~~8095~~	~~8095~~	~~7951~~	~~62~~	~~8106~~	~~8108~~

Table 4. Statistical evaluation of EMAC simulated aerosol concentrations against monthly average observations from East Asia during 2005–2008.

EANET Network

Metric	NO_3^-	Na^+	Ca^{++}	K^+	Mg^{++}	NH_4^+	Cl^-	SO_4^{--}
Observed (µg m^{-3})	1.16	1	0.74	0.36	0.16	1.1	1.39	4.44
Calculated (µg m^{-3})	0.47	0.58	0.21	0.09	0.11	0.44	1.05	1.46
MAGE (µg m^{-3})	0.94	0.79	0.7	0.29	0.14	0.77	1.43	3.18
MB (µg m^{-3})	-0.69	-0.42	-0.53	-0.27	-0.05	-0.66	-0.34	-2.98
NME	0.82	0.79	0.95	0.83	0.83	0.7	1.03	0.72
NMB	-0.59	-0.4	-0.7	-0.75	-0.27	-0.59	-0.21	-0.67
RMSE (µg m^{-3})	2.24	1.53	1.83	0.7	0.22	1.54	2.59	5.02
number of comparisons	1279	1274	1528	1523	1414	1277	1140	1294

Table 5. Calculated average tropospheric burden of inorganic components in the base case and the sensitivity simulations

	Tropospheric Burden of Inorganic components (Tg)										
Simulation Case	NO_3^-	~~Total~~ HNO_3 + NO_3^-	Dust	Na^+	Ca^{++}	K^+	Mg^{++}	NH_4^+	~~Total~~ NH_3 + NH_4^+	Cl^-	SO_4^{--}
1. Base case[1]	0.45	2.10	32.90	3.54	4.70	1.94	1.78	0.17	0.99	3.50	1.78
2. ~~No active dust~~Chemically inert dust	0.25	2.17	38.21	2.02	0.08	0.07	0.25	0.29	0.80	3.20	1.65
3. Homogeneous size distribution of dust	0.41	2.12	20.56	2.87	2.83	1.22	1.22	0.18	0.97	3.45	1.80
4. Uniform chemical composition of dust	0.38	2.14	35.34	2.95	1.88	1.20	0.92	0.19	0.96	3.43	1.77
5. 50% ~~reduced~~ decrease in dust emissions	0.41	2.13	18.61	2.90	2.69	1.13	1.12	0.19	0.97	3.46	1.82
6. ~~Metastable state case for aerosols~~Aerosols in a metastable state	0.44	2.09	32.13	3.54	4.57	1.89	1.74	0.17	0.99	3.50	1.78

[1] The basecase simulation takes into account the chemically active mineral dust components and it assumes that aerosols can form solids (stable state). The emission inventory used includes an explicit geographical representation of the emitted dust particle size distribution and chemical composition.

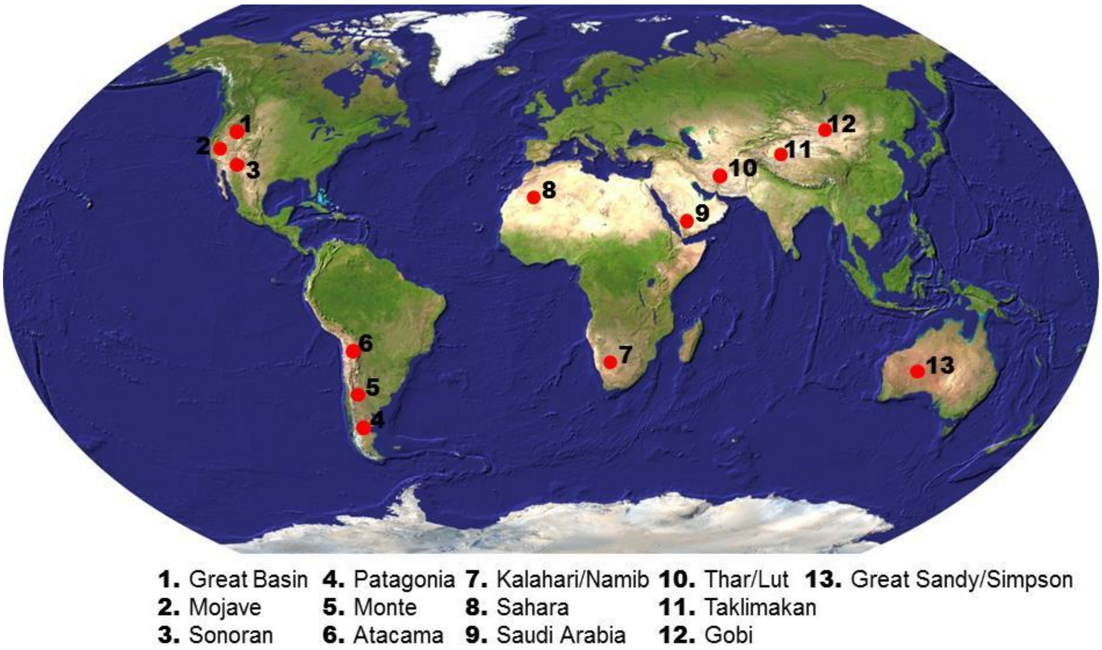


Figure 1: Location of the main desserts of the world in which a discrete chemical composition of the emitted mineral dust is used.

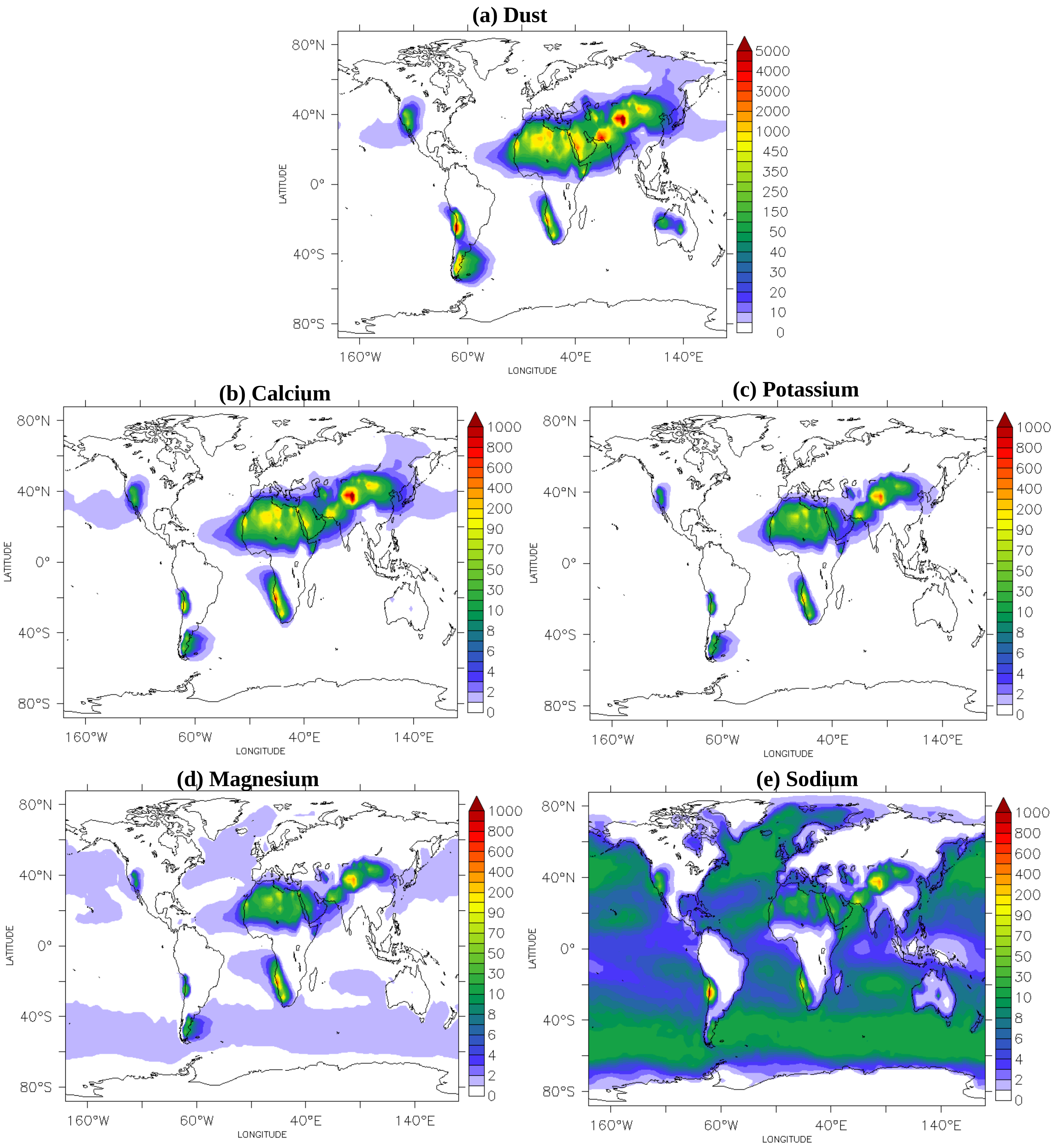
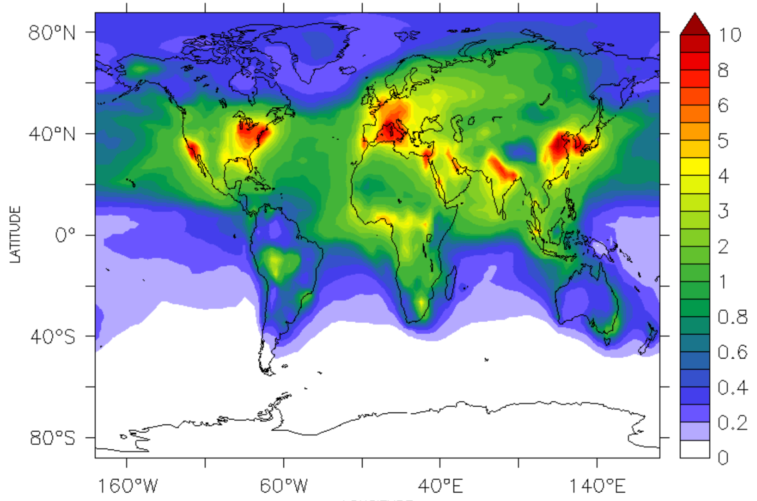
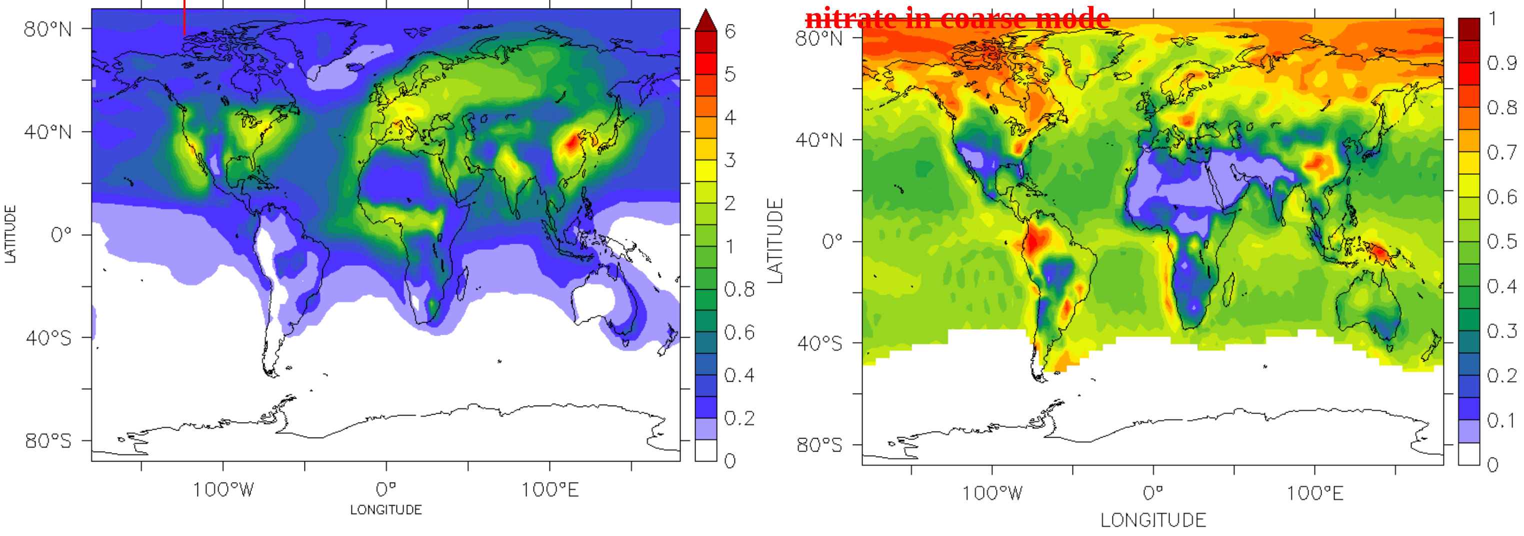


Figure 2~~1~~: Predicted average near-surface concentrations (in µg m^{-3}) of **(a)** inert dust, **(b)** calcium, **(c)** potassium, **(d)** magnesium and **(e)** sodium during the years 2005-2008.



(a) Total nitrate (gas plus aerosols)

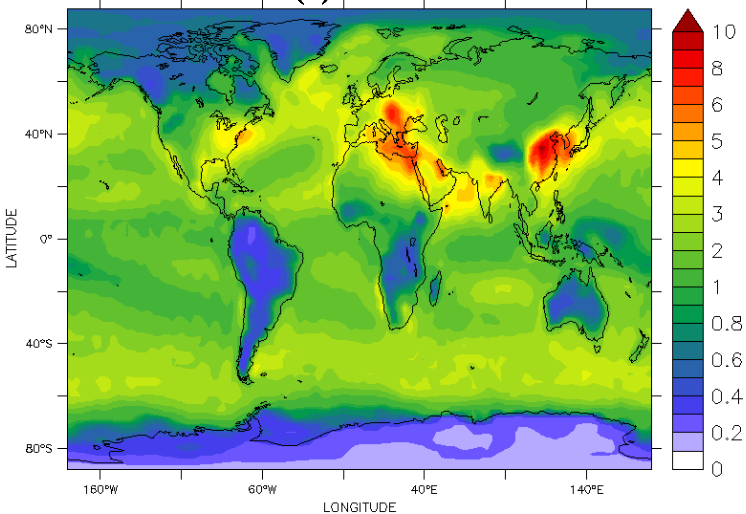


(b) Aerosol nitrate ~~in fine mode~~

(c) Fraction of fine mode aerosol nitrate~~Aerosol nitrate in coarse mode~~

Figure 3~~2~~**:** Predicted average near-surface concentrations (in µg m^{-3}) of **(a)** total nitrate (sum of gas and aerosol phases~~),~~) and **(b)** aerosol nitrate ~~aerosol~~, and ~~in fine mode and~~ **(c)** fraction of fine mode aerosol nitrate ~~aerosol in coarse mode~~to total aerosol nitrate during the years 2005-2008.

(a) Sulfate



(b) Ammonium **(c) Chloride**

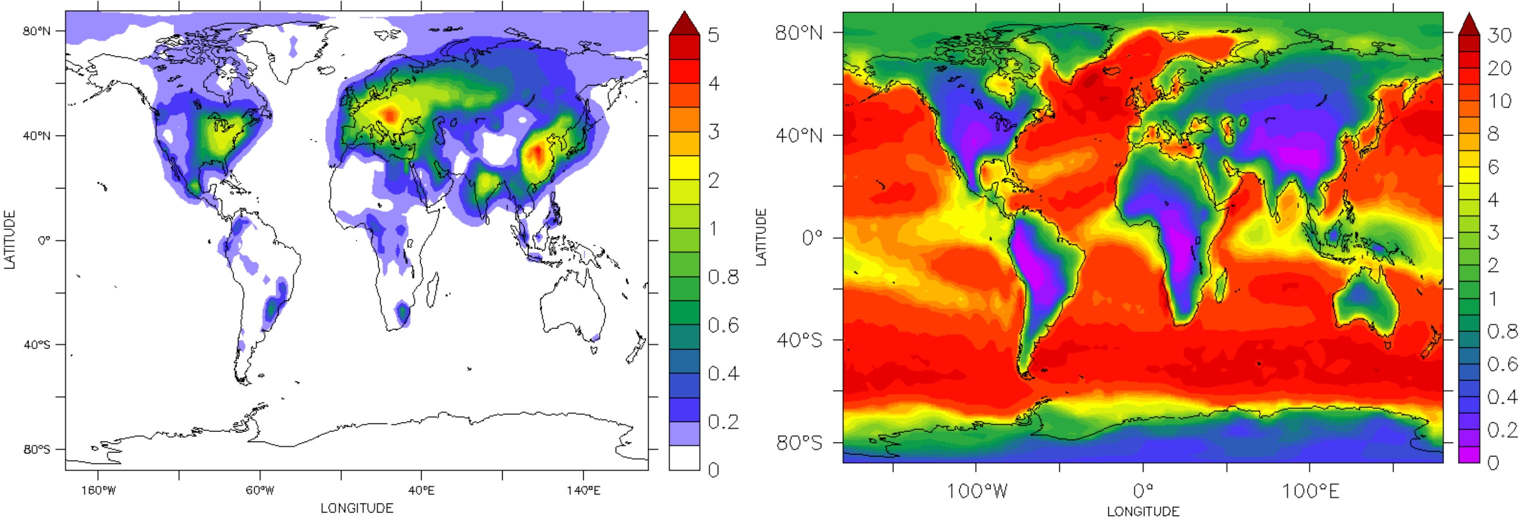
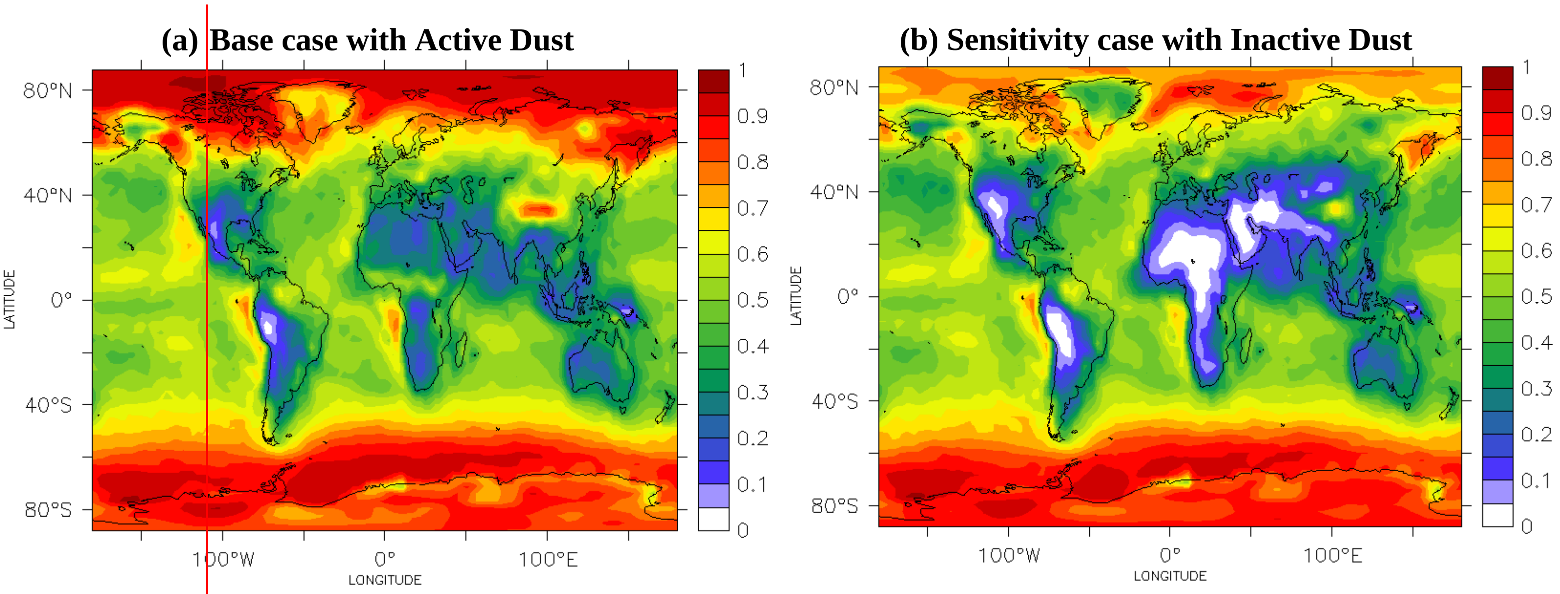


Figure 4~~3~~: Predicted average near-surface concentrations (in µg m^{-3}) of **(a)** sulfate, **(b)** ammonium and **(c)** chloride during the years 2005-2008.



(a) Base case with Active Dust

(b) Sensitivity case with Inactive Dust

~~**Figure 4:** Predicted average near-surface fraction of total nitrate (gas plus aerosol) in the aerosol phase **(a)** by simulating the effect of mineral dust components on its formation (base case), and **(b)** by ignoring the presence of the reactive dust components (sensitivity case) during the years 2005-2008.~~

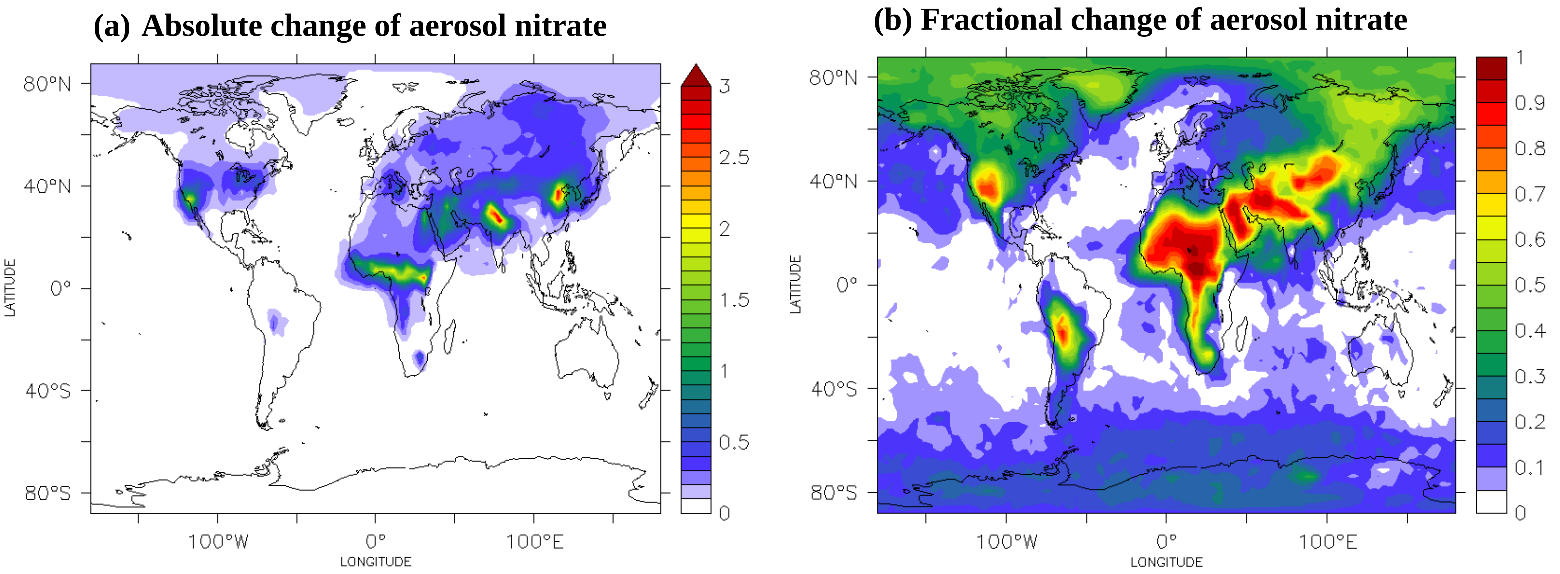
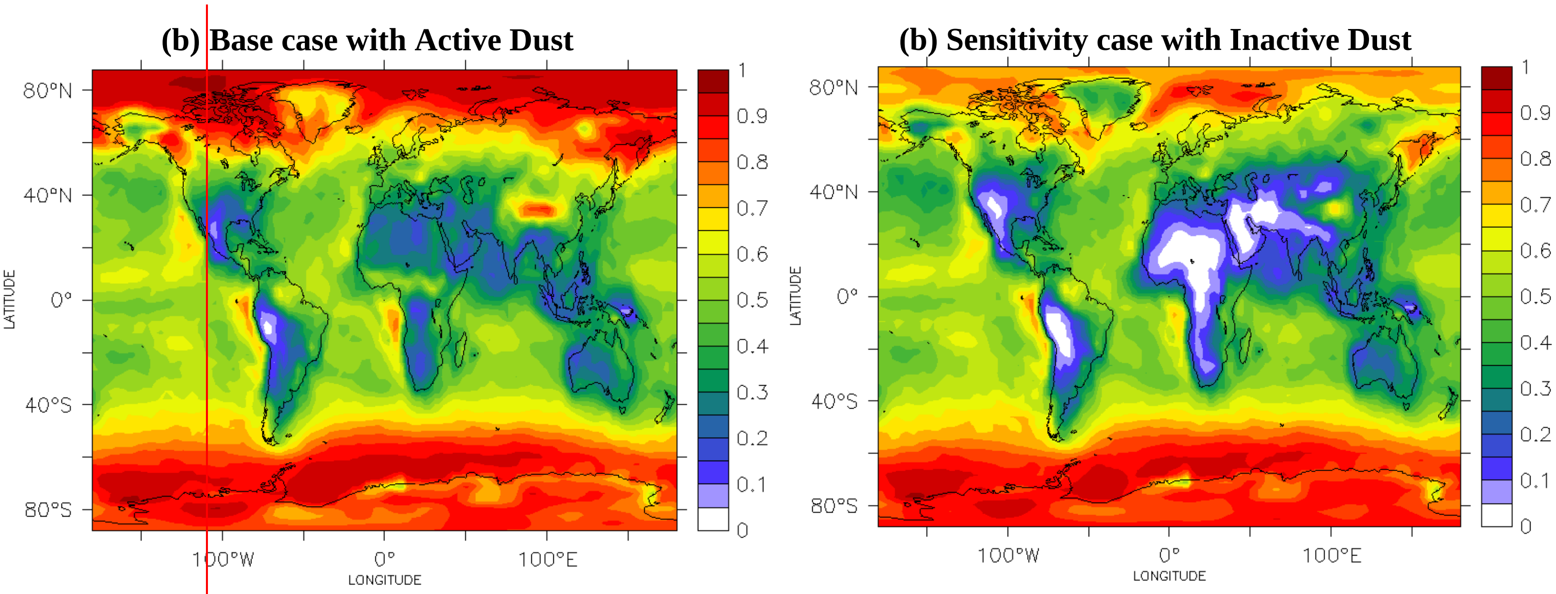


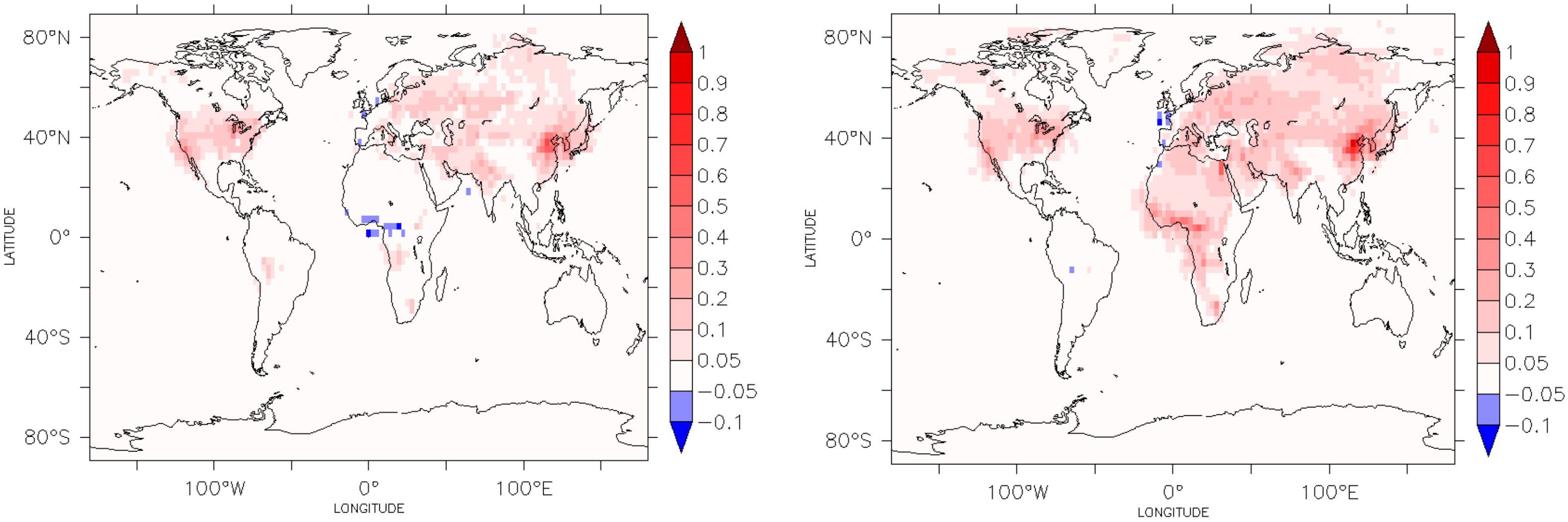
Figure 5: **(a)** Absolute (in μg m^{-3}) and **(b)** fractional change of the predicted average near-surface aerosol nitrate by ignoring the effect of the reactive dust components on its formation, during the years 2005-2008. A positive change corresponds to a decrease after assuming chemically inert dust.



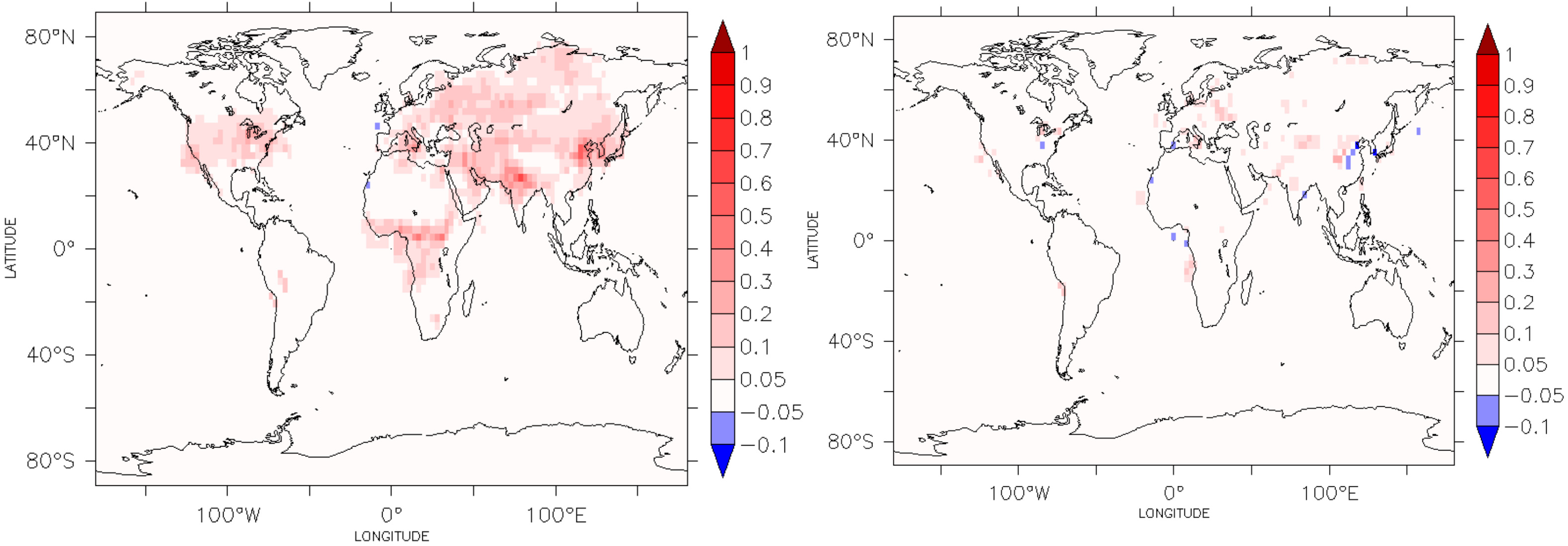
(b) Base case with Active Dust

(b) Sensitivity case with Inactive Dust

Figure 6: Predicted average near-surface fraction of total nitrate (gas plus aerosol) in the aerosol phase **(a)** by simulating the heterogeneous chemistry between dust and nitrate species (base case), and **(b)** by switching off dust chemistry (sensitivity case) during the years 2005-2008.


(a) Effect of the dust size distribution

(b) Effect of the dust chemical composition


(c) Effect of the emitted dust aerosol load

(d) Effect of the aerosol state assumption

Figure 7~~6~~: Absolute changes (in μg m^{-3}) of the predicted average near-surface aerosol nitrate after **(a)** using a dust emission parameterization scheme that utilizes a homogeneous global soil size distribution of dust particles, **(b)** assuming a global uniform chemical composition of mineral dust, **(c)** a 50% reduction of mineral dust emissions and, **(d)** assuming metastable state for aerosols, during the years 2005-2008. A positive change corresponds to a decrease in the sensitivity simulations.