

## REFEREE #3

### Summary

5 Nitrogen is important in several perspectives, and it can affect air quality mainly through oxidized and reduced nitrogen. This study focuses on the nitrate formation processes, and designed a series of numerical experiments to elucidate the main governing mechanisms. The overall study is important and interesting, but I do feel the manuscript needs a substantial revision to be easily followed.

10 We appreciate the reviewer's comprehensive revision and insightful comments, which we think point to important aspects of the paper that deserve attention. We address the comments in the lines below. Additionally, the original text of the manuscript has been substantially revised to ease its readability and comprehension, as outlined in the responses that follow.

### Comments

15 1. Abstract: regarding the first sentence. I am not sure the main focus is nitrate or the desert dust. I feel the authors try to elucidate the issues of nitrate, and dust is one of the factors affecting concentrations of nitrate. Line 5-6: "This study investigates key processes in nitrate formation over dust and evaluates their representation in models." By reading the manuscript, I think the authors not only examine nitrate formation over dust, but also on sea salt and others. Does the abstract correctly deliver the message?

20 We understand this comment and it was indeed a topic of discussion among the authors of the manuscript.

While our study incorporates nitrate formation over sea salt and other aerosol species, these are included primarily because they represent crucial pathways to accurately reproduce global particulate nitrate observations. However, the main scope of the work is to investigate the role of dust and its representation in models for nitrate formation. Through systematic sensitivity simulations, we aim to better understand the pathways driving nitrate formation on dust. Some sensitivity simulations - such as the inclusion or exclusion of nitrate formation on sea salt in some mechanisms - are intended to evaluate the role of sea salt in the global nitrate burden, given its competition with dust for gas-phase species. However, these sensitivity simulations do not aim at specifically investigating the mechanisms governing nitrate formation on sea salt.

25 30 For these reasons, we emphasize the role of dust representation in the formation of nitrate in several parts of the manuscript, including the abstract, instead of referring to a more general nitrate formation analysis and discussion. The scope of the work is better clarified in the revised manuscript.

35 2. Introduction: the authors have tried to discuss the potential issues related to heterogeneous reactions (e.g.,  $\text{HNO}_{3(g)}$  to dust). Similar as the previous question: I am not sure the main focus of this study is dust or nitrate. I feel it is nitrate instead of desert dust. At the third last paragraph, the authors pointed out a general inaccuracy of current models in reproducing nitrate and misrepresentation of nitrogen heterogeneous chemistry processes on dust and sea salt. Following this, the authors mentioned Following that, the authors try to systematically investigate the underlying processes governing the issues. However, I feel it is not clear what issues are in the current models. Is it due to the DMT or TEQ? It does not look like the authors have discussed the problems clearly. In this way, the readers would not know how the authors can disentangle the problems.

40 We understand and appreciate the reviewer's concern regarding the potential ambiguity in the scope of the present study. As clarified in the previous comment, our study aims to study the mechanisms governing nitrate formation on mineral dust, assessing its sensitivity to critical variations in nitrate formation pathways on these species. While some sensitivity simulations intend to assess the relative role of, for instance, sea salt, they do not explore the mechanisms governing the formation on other

45 species.

Regarding the clarity in the presentation of the issues faced by current models, we appreciate the reviewer's perspective on this matter.

50 For this study we conducted a comprehensive literature review to understand and contextualize existing dust chemistry mechanisms incorporated in atmospheric models, as it is included in the introduction section. This review served to understand that very often, difficulties faced by models when reproducing global particulate nitrate observations could not be attributed solely to their heterogeneous dust chemical mechanism (i.e. TEQ, HYB, DBCLL or DMT), but also to a wide variety of other factors, such as the emission inventories, the alkalinity representation of dust and sea-salt or their size distribution parameterization, among many others. Indeed, the AeroCom Phase III nitrate experiment (Bian et al., 2017) highlights the significant  
55 disparities among participating models, driven by differences in the factors mentioned.

In this study, we conducted a systematic investigation of the key chemical factors influencing particulate nitrate formation on dust, utilizing a single atmospheric chemistry model with consistent transport, emissions, and chemical schemes across all experiments. Therefore, we do not delve into the specific issues faced by current models when trying to reproduce nitrate formation on dust, leaving the possibility to, in future studies, investigate other factors such as variations in dust mineralogy.  
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We have clarified the scope of our work in the Introduction (line 106 of the revised manuscript):

"The scope of the present work is to understand the role of dust in  $\text{NO}_3^-$  formation through a systematic investigation of the underlying processes governing dust heterogeneous chemistry. [...] While our primary emphasis is on the heterogeneous chemistry on dust surfaces, we also account for nitrate formation on SS and its alkalinity."  
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3. Line 154: "assuming complete nucleation" I am not sure what complete nucleation means. Does this mean that all gas  $\text{H}_2\text{SO}_{4(g)}$  is nucleated? This does not seem reasonable.

70 By "complete nucleation", we mean that all gas  $\text{H}_2\text{SO}_{4(g)}$  remaining in the atmosphere after aqueous sulfate formation is assumed to partition to the aerosol phase through nucleation, as pointed out by the reviewer.

The assumption of complete nucleation for  $\text{H}_2\text{SO}_{4(g)}$  is a simplification to represent  $\text{H}_2\text{SO}_{4(g)}$  transfer to particulate phase in a mass-based model such as MONARCH. As the model does not include particle microphysics nor operates as a number-concentration based model, this approach ensures a first order treatment of nucleation and condensation processes of  $\text{H}_2\text{SO}_{4(g)}$   
75 in the model. The products of both processes are treated equally in the model, both represented within the model's fine particulate sulfate bin (Spada, 2015). Given the extremely low vapor pressure of  $\text{H}_2\text{SO}_{4(g)}$  (Yu and Luo, 2009; Wang et al., 2015), it is reasonable to assume the complete partitioning of  $\text{H}_2\text{SO}_{4(g)}$  into the particulate phase through nucleation and condensation. Similar approximations have also been assumed in other models, such as GoCart (Chin et al., 2000) and IFS-AER (Rémy et al., 2019), where even a more simplistic approximation is considered to account for sulfate mass production in the  
80 atmosphere through a single kinetic reaction oxidizing  $\text{SO}_{2(g)}$  and directly forming particulate sulfate.

To clarify the concept of complete nucleation, we introduced the following text in Section 2.1 of the revised manuscript (line 159):

"At the end of each chemistry integration time step, the remaining  $\text{H}_2\text{SO}_{4(g)}$  that has not formed aqueous sulfate is assumed to fully nucleate into fine particulate  $\text{SO}_4^{2-}$  (Spada, 2015)."  
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Although the assumption of complete nucleation of  $\text{H}_2\text{SO}_{4(g)}$  may not be considered completely accurate, in our particular context of modeling dust heterogeneous chemistry, the phase of  $\text{H}_2\text{SO}_{4(g)}$  (either solid, liquid or gas) does not influence the particulate nitrate and ammonium formation from the thermodynamic equilibrium calculation. This is because the ISORROPIA-II thermodynamic model uses total sulfate - indistinctively of its phase - as input to compute the nitrate partitioning. Furthermore,  
90 since in our work we assume metastable conditions with ISORROPIA-II, it does not consider the precipitation of solid particles from  $\text{H}_2\text{SO}_{4(g)}$  condensation. Consequently, the thermodynamic equilibrium calculation does not affect the partitioning of  $\text{H}_2\text{SO}_{4(g)}$ , and we assume its complete condensation into the respective size mode. This consideration would change if we

employed ISORROPIA-II in the stable mode, where solids from the partitioning of  $\text{H}_2\text{SO}_4(\text{g})$  (such as  $\text{CaSO}_4$ ) would need to be explicitly accounted as outputs.

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4. Line 181: “In this study, we investigate the primary chemical pathways responsible for  $\text{NO}_3^-$  formation on coarse particles by integrating” Why do the authors only investigate the pathways of nitrate formation on coarse particles? How about the fine mode?

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We concur with the reviewer in the possible confusion that this sentence might lead to. We modified the sentence to try to improve its clarity to (Section 2.2 line 188 of the revised manuscript):

“In this study, we investigate the primary chemical pathways responsible for  $\text{NO}_3^-$  formation on preexisting particles, with a particular focus on coarse dust particles, by integrating mechanisms of varying complexity within the global model MONARCH”

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We consider it is important to note that particular focus is put in the role of “coarse particles” since most of the sensitivity tests are performed varying the mechanisms for coarse nitrate formation particularly on coarse dust particles. Conversely, fine nitrate formation is consistently formed through TEQ in all mechanisms analyzed here.

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5. Line 188 hydrophilic Why hydrophilic? Some explanation is needed.

We agree in that the text should provide a better explanation of this point. The additional bin is considered to be hydrophilic because the studied formation mechanisms occur under moist conditions, a regime in which the nitrate formation pathways are more efficient. For instance, the uptake coefficient on dust as a function of relative humidity is parameterized for non-zero humidity values. Additionally, thermodynamic equilibrium is performed under the metastable solution assumption (i.e. considering only gas and liquid species). Therefore it requires a positive relative humidity to account for the deliquescence of crustal species.

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We added a clarification as follows (Section 2.2 line 193 in the revised manuscript):

“To trace the formation of fine and coarse  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{SO}_4^{2-}$  under moist conditions — the primary regime where these formation pathways occur (Usher et al., 2003; Jordan et al., 2003) — an additional hydrophilic bin for the coarse mode of these species is added to the default MONARCH size parametrization, as detailed in Supplementary Table S2.”

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6. Table 1: Not sure why in this table, there is no uptake of  $\text{HNO}_3(\text{g})$  on dust. Indeed, Line 214: the authors mentioned that “For the uptake of  $\text{HNO}_3(\text{g})$  on dust (R4-5 in Table 1)”. I don’t understand why R5 is related to dust (it apparently says sea salt). The caption of Table 1 says DU represents Dust, why specifically using  $\text{CaCO}_3$ ?

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We thank the reviewer for highlighting this point. The sentence in line 214 has a typo and should only refer to reaction R4. We have amended the sentence in the revised manuscript.

In Table 1, the uptake of  $\text{HNO}_3(\text{g})$  on dust is expressed in R4. The explanation why we used  $\text{CaCO}_3$  instead of Dust is addressed in the note 3 of Table 1. For this reaction, we scale the dust uptake coefficient to the actual  $\text{CaCO}_3$  content to account for alkalinity. Consequently, the uptake is considered to happen only on  $\text{CaCO}_3$  particles. This is not the case for reactions R1 and R3. In the case of  $\text{SO}_2(\text{g})$  (R3), the uptake coefficient is also scaled for  $\text{CaCO}_3$  content with  $Sc$ . However, an exception is made here, setting  $Sc$  to 1.0 in the case without alkalinity. That is why in Table 1 reaction R3, DU is specified instead of  $\text{CaCO}_3$ . This was mentioned in Note 2 of Table 1, but we clarify the explanation of the note as follows:

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“That is why this reaction is considered to happen over DU(aer) and not only over  $\text{CaCO}_3(\text{aq})$ , although it assumes same dust alkalinity as in nitric acid uptake (R4).

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145 7. Line 216-218: “However, literature reports varying values for  $Sc$  based on dust alkalinity assumptions, ranging from  $Sc=1/30$  for the industrially-standardized Arizona Test Dust (Möhler et al., 2006; Herich et al., 2009; Suman et al., 2024) to  $Sc=0.018$  for samples from the China Loess (with 39%  $CaCO_3$  content) (Krueger et al., 2004; Wei, 2010).” What is the setting in this study considering the large range of  $Sc$ ?

150 The values adopted for  $Sc$  in our study are specified in Line 224 of the original manuscript: “ $Sc = 1.80$  and  $Sc = 1.52$  for the two alkalinity values used in our experiments”. It is important to note that we use the alkalinity scaling factor  $Sc$  to normalize the Fairlie et al. (2010) values, which assumes a different non-volatile cation content than our work. This is explained in Lines 219-225 in the original manuscript. We did not evaluate alternative  $Sc$  values, given that our approach considers a constant dust mineralogical composition.

155 8. For  $RH$  above 90%,  $\gamma(SO_{2(g)})$  remains constant at  $\gamma(SO_{2(g)}) = Sc \cdot 5.0 \cdot 10^{-4}$  What is the value of  $Sc$  in this equation

In this case,  $Sc$  has the same value as the  $HNO_{3(g)}$  case,  $Sc = 1.80$  for Journet et al. (2014) and  $Sc = 1.52$  for Claquin et al. (1999) mineral datasets, as mentioned in Lines 224 and 229 and Section 2.2.1 in the original manuscript.

However, the uptake reaction of  $SO_{2(g)}$  is performed even in the absence of alkalinity, where  $Sc$  is set to 1.0 and not 0.0, as it was assumed for  $HNO_{3(g)}$  uptake in the absence of alkalinity (see note 2 in Table 1).

160 We clarified the original sentence in the revised manuscript as follows (Section 2.2.1 line 241):

“The same  $Sc$  values as used for  $\gamma(HNO_3)$  are applied for  $\gamma(SO_2)$ :  $Sc = 1.80$  and  $Sc = 1.52$ . However, if alkalinity is not considered,  $Sc$  is set to 1.0 (and not zero, as the case for  $\gamma(HNO_3)$ ) to account for the oxidation of  $SO_{2(g)}$  by deliquesced  $O_3$  and  $NO_2$  (Usher et al., 2002; Prince et al., 2007; Yu et al., 2017; Li et al., 2024). For  $RH$  above 90%,  $\gamma(SO_2)$  remains constant at  $\gamma(SO_2) = Sc \cdot 5.0 \cdot 10^{-4}$ .”

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9. Line 301: The dust NVC global average content result in: 5.17%  $Ca^{2+}$ , 0.79%  $Na^+$ , 2.37%  $K^+$ , 1.32%  $Mg^{2+}$  for the Journet et al. (2014) dataset, and 3.68%  $Ca^{2+}$ , 0.87%  $Na^+$ , 3.15%  $K^+$ , 1.75%  $Mg^{2+}$  for Claquin et al. (1999). Which one does the author use? Please add some descriptions besides of laying out the two different values.

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Table 3 column DU Alkalinity specifies which dataset is used in the different sensitivity runs discussed in our work. Global average values derived from Journet et al. (2014) are employed in all runs except the case DBCLL\_ClaqAlk, where Claquin et al. (1999) dataset is used. With the exception of the DBCLL\_ClaqAlk run, all the others experiments use 5.17%  $Ca^{2+}$ , 0.79%  $Na^+$ , 2.37%  $K^+$ , 1.32%  $Mg^{2+}$  NVC percentages, derived from Journet et al. (2014).

175 This is now clarified in the revised manuscript Section 2.2.3 lines 319-323 adding a brief description of the main differences between the two datasets as follows:

“In most of the sensitivity runs, the Journet et al. (2014) global average is employed, if not stated otherwise (see Table ??). Values for Claquin et al. (1999) are used solely in one sensitivity test, as explained in Section ??. These values are within the range reported by Karydis et al. (2016) ( $5.36 \pm 3.69\%$   $Ca^{2+}$ ,  $2.46 \pm 1.90\%$   $Na^+$ ,  $2.08 \pm 1.34\%$   $K^+$ ,  $1.96 \pm 2.20\%$   $Mg^{2+}$ ). The Journet et al. (2014) dataset results in a higher proportion of  $Ca^{2+}$  compared with Claquin et al. (1999), while similar fractions for  $Na^+$  and  $K^+$  are reported.”

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185 10. Table 4: there is no unit

We thank the reviewer for pointing out the missing units in Table 4. We have amended this by specifying the units in the Table’s caption. Both bias and rmse are reported in  $\mu g m^{-3}$ .

190 *11. Descriptions in section 3: I feel it is not easy to follow. Too many numbers. Too many abbreviations make it very hard to follow. If possible, please elaborate a bit more to emphasize the main focus. More references can be added to enhance the readability, and readers might know what is important.*

We value this observation and agree with this comment. The text of this section has been significantly revised and reduced to ease the reading and highlighting the main scope and findings of our results. We simplified the definitions, the use of abbreviations, and the numerical results provided in the original manuscript that were not relevant for the main findings of each subsection. We also moved some experiments' analyses to the Supplementary material (Section S5). We believe that the revised section addresses now the concerns raised by the reviewer.

200 *12. The authors mentioned many times about dust and sea salt. Did the authors do any evaluation on these two species? Can the model reasonably reproduce dust and sea salt?*

The present work does not specifically evaluate dust nor sea-salt concentrations. For dust, this study adopts the same scheme evaluated in Klose et al. (2021) and Gonçalves Ageitos et al. (2023), in both studies a comprehensive evaluation of the dust cycle were provided. Concerning the sea-salt, our simulations used the scheme already evaluated in Spada et al. (2013) where different parameterizations were compared and extensively evaluated. In our work, we report dust and sea-salt column loads in Supplement Figures S10 and S11 to support the discussion on nitrate formation and focus the main evaluation to nitrate and particulate matter.

210 To address the reviewer comment, we have added the following sentence on the assessment of the dust and sea salt life cycle simulated by the model citing the mentioned references (Section 2.1 lines 185-186 of the revised manuscript):  
"For a detailed evaluation of the model's dust cycle, the reader is referred to Klose et al. (2021) and Gonçalves Ageitos et al. (2023), and for SS to Spada et al. (2013)."

215 *13. For the reduced and oxidized nitrogen, did the authors compare the results with observations? I think comparison with observations are useful to understand the nitrogen budget.*

We value this recommendation and agree that this evaluation would be useful. Consequently, we added the evaluation of reduced and oxidized species in the Supplementary Section S6.

225 The discussion of the evaluation is also included now in Section 3.3.2 (lines 844-848 of the revised manuscript) as follows:  
"The observational evaluation of both reduced and oxidized species, along with the monitoring stations used, can be found in the Supplementary Section S6. Overall, a general good agreement with observations is obtained. Oxidized nitrogen species exhibit low biases in all mechanisms except HYB\_du-ssUPTK, which overestimates observations throughout the studied period, supporting the conclusion that UPTK coefficients used are excessively efficient. Reduced nitrogen species, while slightly underestimated, remain well within the observational variability range."

230 *14. The conclusions: It seems not clear what is the best option to take for the nitrate reactions. The reversible or irreversible? Please summarize to make it clear whether there is an optimal option, or multiple options to derive reasonable simulations of nitrate.*

We thank the reviewer for pointing this out and agree that our manuscript did not provide a clear recommendation regarding the most suitable scheme for simulating nitrate formation. Based on our analysis and evaluations, we recommend adopting the reversible representation in models, as it enables a more detailed description of key processes, such as alkalinity in nitrate partitioning, without significantly increasing the computational cost.

We have introduced a statement in the Conclusions (line 1030) to highlight the best mechanism based on our analysis as follows:

240 “Overall, the DBCLL<sub>du-ssAlk</sub> scheme demonstrates the best accuracy compared to the other tested configurations, as evidenced by its closer alignment with observations.”

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