

I thank authors for responding to both reviewers. I have some comments regarding their responses and some additional remarks that need to be considered before publication.

### **Concerning responses to R1:**

R1 comment #1: “They attribute these model differences to differences in 1) pH-dependent wet deposition of  $\text{NH}_4^+$ , 2) nitrate formation on the surface of sea salt and dust aerosol, and 3) the nitrate coarse mode fraction. They find that nitrate production on sea salt and dust is important to include in models as it tends to dominate nitrate production and controls its partitioning between the fine and coarse mode. In that sense, it seems to me that 2 and 3 above are referring to the same process.”

Author’s response: “We intend to separate discussion of section 5.2 and 5.3 because the nitrate formation on the surface of sea salt and dust aerosol (section 5.3) is important, but not the only factor, to determining nitrate size distribution (section 5.2). Also, the former focuses more on chemical process and the later on physical process and climate implication.” I find that the authors did not address the reviewer’s concern. The formation of nitrate on coarse mode dust and sea salt particles is the major factor controlling the size distribution and the ratio of coarse/total particulate nitrate in models. This is not clearly explained in the manuscript, and should be clarified in section 5.

R1 comment #2: Authors did not address the reviewer’s concern “What the authors are referring to by the use of “heterogeneous chemistry” is what I would call thermodynamic partitioning between the gas and aerosol phase.” I agree with the reviewer that the use of the heterogeneous chemistry should not be applied to refer to the gas-particle thermodynamic partitioning. It is unclear in the revised manuscript whether the models are treating the formation of coarse mode nitrate by a heterogeneous uptake of  $\text{HNO}_3$  onto dust and sea-salt particles or by an equilibrium approach. This must be clarified.

### **Concerning responses to R2:**

In the added text please change the 1<sup>st</sup> sentence to: “Our work presents an initial effort to assess nitrate simulation from chemical and physical processes (deposition).” You should be more specific by what chemical and physical processes you have looked at. And please have the rest of the paragraph corrected for English.

Why would you want to compromise the accuracy and efficiency, this sentence is misleading (line 619: “Several approximations, therefore, have been developed to compromise accuracy and efficiency.”) Is this what you meant: “Several approximations have been developed to allow computational efficiency although they might compromise the model accuracy.” Please reword.

### **Additional comments:**

1) The confusion about the use of the “nitrate” term:

It should be clearly stated in the title that the paper is evaluating the particulate nitrate: “Investigation of global particulate nitrate from the AeroCom Phase III experiment.”

Instead of using nitrate aerosol it would be preferable to use particulate nitrate when referring to the particle phase as aerosol term refers to both gas and particulate fraction that are in equilibrium.

Line 122: Is this particulate or gas-phase nitrate: “If fixed Nr is deposited as nitrate in forests,..”

2) As mentioned by R1, N<sub>2</sub>O<sub>5</sub> hydrolysis is an important heterogeneous reaction when investigating the nitrate budgets that is typically included in global models. It should be clearly stated in Table 1 or 2 and in the paper how this reaction is treated and if it is included. And some discussion on the uncertainty due to this reaction and references should be added in the manuscript.

3) Add “relative” Line 78: More importantly, the relative importance of aerosol nitrate

4) Add “particulate” Line 102: First, the formation of particulate nitrate,

5) Given that coarse mode nitrate measurements are sparse, please include measurements that have been done in Paris during the ESQUIF campaign that found that the coarse nitrate fraction represents up to 60% of the total particulate nitrate mass during the night and 80% during the day. See either Figure 13c of “Hodzic et al., ACP 2006 Aerosol chemical and optical properties over the Paris area within ESQUIF project”, or Figure 6 of Hodzic et al, AE 2006: A model evaluation of coarse-mode nitrate heterogeneous formation on dust particles.”

6) Clarify what you mean by feedback in this sentence: Line 202: “All models use full gas phase O<sub>3</sub>-NO<sub>x</sub>-HO<sub>x</sub> chemistry to produce HNO<sub>3</sub> and consider the feedback of nitrate aerosol formation on HNO<sub>3</sub> calculation.” Do you mean radiative feedbacks on photolysis or changes in the HNO<sub>3</sub> concentrations due to the gas/particle equilibrium?