EGUsphere, referee comment RC1
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Comment on egusphere-2022-178

Anonymous Referee #1

Referee comment on "A comprehensive study about the in-cloud processing of nitrate through coupled measurements of individual cloud residuals and cloud water" by Guohua Zhang et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-178-RC1, 2022

Review comments for Zhang et al

This paper aims to investigate the formation and evolution of nitrate in clouds, which has been rarely studied, based on the size-resolved mixing state of nitrate in the individual cloud residual and cloud-free particles by single particle mass spectrometry and the mass concentrations of nitrate in the cloud water and PM2.5 in southern China. The data show the direct observational evidence for the enhanced nitrate formation in the cloud water and residual particles, which is attributed to the enhanced hydrolysis of N2O5. Such a comprehensive dataset is quite robust for discussions and conclusions. Given that the in-cloud process is critical to accurately evaluating the evolution and oxidative impacts of nitrate, which is increasingly important, the manuscript is worthy of publication after considering my suggestions.

Major Comments:

(1) The authors first exclude the scavenging of gas-phase HNO3 as a major pathway through the analysis of the size distribution of nitrate RPA and RPA ratio (nitrate/sulfate). However, the discussions based on such data are not clear enough. More detailed comparison and/or theoretical basis should be included to support the discussion.

(2) The authors show that the hydrolysis of N2O5 explains ~1-3% increase in the nitrate mass fraction in clouds, whereas the in-cloud processing contributed to > 5% increase, and two possible explanations were provided. I wonder if it is possible to reveal the most important factor and if there are other possibilities since the consideration of these explanations may still not fully explain the observation.

(3) In Fig. S1, I noticed that the relative humidity is close to 100% during cloud events, but RH inevitably declines during the cloud-free period. Given that RH has a certain impact on the hydrolysis of N2O5 and the scavenging of gas-phase HNO3, which is worth to have some discussions on it.

Specific comments:

(1) Introduction: The authors summarized the previous studies on the in-cloud processing of nitrate and showed that the detailed observational investigations and the possible mechanisms governing nitrate behavior upon in-cloud processes are scarce. It would be better to include how these studies quantify the relative roles of each pathway for the formation of nitrate in clouds.

(2) Line 141: The predefined sampling cloud droplet size is 7.5-8.5 μm. So why is the mean droplet size assumed to be at 7 μm when calculating the SA?

(3) Line 284: More detailed discussion should be provided to indicate the fast heterogeneous uptake coefficient, and also the specific values.

(4) Line 356: The involvements of VOCs may impact the formation of nitrate through various mechanisms, e.g., direct reaction with NO3 or consumption the oxidants such as OH, which should be discussed in detail.

Figure 3: It will be easier to understand if the chemical formulas N2O5 and NO3- are written in the axes of the figure instead of “nitrate”.

SI line 47: It should be “Text S1”.

SI figure S4: It's difficult to understand this figure, can you describe the horizontal and vertical coordinates more clearly in the figure caption?