Dear Editor and Referee

Thank you for reviewing and commenting upon our manuscript, "Elucidate the Formation Mechanism of Particulate Nitrate Based on Direct Radical Observations in Yangtze River Delta summer 2019" by Tianyu Zhai et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-548-RC1, 2022. As detailed below, the reviewer's comments are in italicized font, and our responses to the comments are in regular font. New or modified text is in blue.

We've responded to each comment individually below and would like to draw your attention to two major concerns:

Comment 1: About Line 75-77 and Line 275-276, the concern about the contribution of NO_2 heterogeneous uptake.

Reply: Thanks for this precious advice. First, we change the description of NO_2 heterogeneous uptake contribution on nitrate formation and add relative literature reports around line 75. What's more, the NO_2 heterogeneous uptake pathway analysis has been added to chapter 3.4. The NO_2 uptake coefficient is set as 5.8×10^{-6} depending on the report by Yu et al. (2021), which is the result of 70% RH on urban grime. The NO_2 heterogeneous uptake pathway contribution on nitrate formation mass increased twice on polluted day than on clean day. However, there remains a distance among the value of NO_2 heterogeneous uptake, $OH+NO_2$, and N_2O_5 heterogeneous uptake. More clarifications have been added in section 3.3 as follows：

As shown in Figure 7c, $OH + NO_2$ dominates nitrate production on clean day, while the N_2O_5 uptake pathway only contributes 13.6 μg m^{-3}. On polluted days, the ability of N_2O_5 uptake grows fast which reached 50.1 μg m^{-3}, while OH pathway doesn't change too much. There is no distinct difference of daytime pathway ($OH + NO_2$) between clean day and polluted day, while the nighttime pathway ratio rises from 38.1 % on clean day to 67.2 % on polluted day.NO_2 heterogeneous uptake increases from 0.93 μg m^{-3} on clean day to 2.0 μg m^{-3} on polluted day, but the contribution proportion do not change obviously. Both the higher N_2O_5 uptake coefficient and higher S_a on polluted day increase the contribution of N_2O_5 hydrolysis on particular nitrate at pollution

condition.

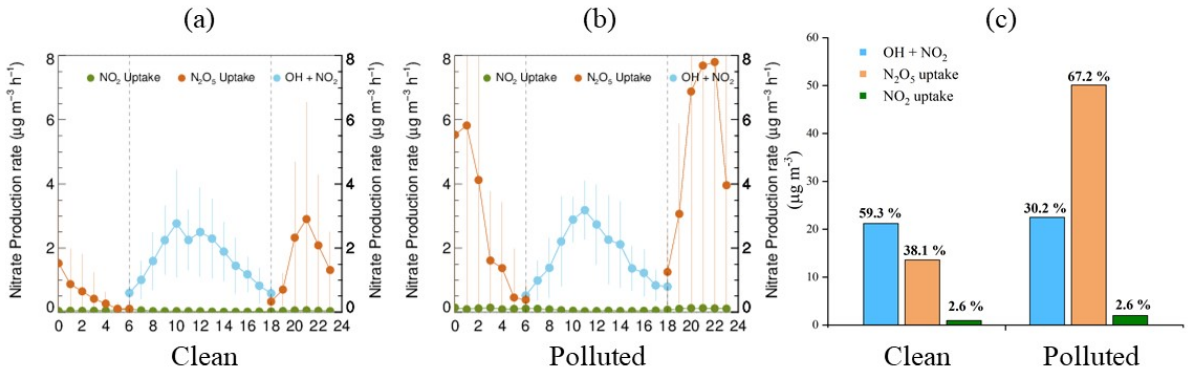

Figure 7 The mean diurnal variations of nitrate production potential of clean day(a) and polluted day (b) and the P(NO_3^-) distribution of clean and polluted day (c).

Comment 2: What does the "water-soluble ion" refer to?

Reply: "water-soluble ion" refer to Na^+, K^+, Ca^{2+}, Mg^{2+}, NH_4^+, NO_3^-, Cl^- and SO_4^{2-} components in particle. In order to avoid confusion, "water-soluble particulate components" is used to replace all the "water-soluble ion".

Comment 3: Can the author elaborate more on the NOR difference between PD and CD? From my perspective, particular factors should lead to such a discrepancy. Any suggestions?

Reply: The NOR increased during PD reveals the fast transformation of NO_2 to NO_3^-, which is in accordance with nitrate ratio explosive growth during PD. As discussed in chapter 3.4, both the N_2O_5 heterogeneous uptake pathway and NO_2 heterogeneous uptake pathway increased more than twice during PD. In our opinion, there is no significant difference in the RH, NO_2 concentration, and N_2O_5 concentration between PD and CD. Even the N_2O_5 concentration decreased during PD night. The nitrate formation contribution increase is controlled by the aerosol surface growth and aerosol water content increase which is due to the change of particulate composition.

Comment 4: Any suggestions on the increased ability of N_2O_5 uptake on a polluted day? Is it

due to the different composition of particles on PD and CD? Need more elaboration.

Reply: Both the higher N_2O_5 uptake coefficient and higher S_a on polluted day increase the contribution of N_2O_5 hydrolysis on particular nitrate at pollution condition. Both the N_2O_5 uptake coefficient and S_a shows a good correlation to RH and aerosol water content. For the N_2O_5 uptake coefficient, although particulate nitrate mass concentration increased during pollution event, antagonistic effect on N_2O_5 uptake coefficient was not obvious for the nitrate molarity decreasing.

Thank you again for your thoughtful comments.

Reference

Yu, C. A., Wang, Z., Ma, Q. X., Xue, L. K., George, C., and Wang, T.: Measurement of heterogeneous uptake of NO2 on inorganic particles, sea water and urban grime, J. Environ. Sci., 106, 124-135, 10.1016/j.jes.2021.01.018, 2021.