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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Two years online measurement of fine particulate nitrate in western Yangtze River Delta: Influences of thermodynamics and $N_2O_5$ hydrolysis" *by* Peng Sun et al.

**Anonymous Referee #2**

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The authors present two-years measurements of fine nitrate aerosol at a rural site in the Yangtze River Delta, China. The data are analyzed to illustrate the seasonal and diurnal variations of particulate nitrate and its formation pathways. It was found that photochemical formation of nitric acid and its thermodynamic equilibrium with NO3- play a dominant role in summer, whilst hydrolysis of N2O5 dominates in winter. Overall, this study provides valuable observational data and useful insights into the chemical behaviors of nitrate aerosol in the polluted atmospheres of China. Thus this manuscript can be accepted for publication after the following comments are properly addressed.



Specific comments:

The major concern is on the steady state calculation of N2O5 and its contribution to the NO3- formation. First, is the steady state assumption valid in this study, especially for the cold conditions in winter? The authors need estimate the chemical lifetimes of N2O5 for the selected cases and examine if the air masses were in steady state for N2O5? Some parameters (e.g., uptake coefficient of N2O5 onto particles) are highly uncertain, which may introduce large uncertainty to the calculation. The chemical loss of NO3 radical via reaction with VOCs is also highly variable and depends on the abundances and chemical speciation of VOCs, especially biogenic VOCs. The authors may conduct more calculations with varying levels of uptake coefficients and reaction rates of NO3+VOCs, to examine the sensitivity of the major conclusions to these assumptions.

Section 3.1: although this manuscript focused on fine particulate nitrate, it should be useful to document the overall measurement results of other related species, such as sulfate, PM2.5, NOx, O3 and NH3. Besides the ratio of nitrate to water-soluble ions, it is also very useful to show the mass ratio of nitrate to PM2.5.

Pg 3, Lines 64-66: the following recent observational studies of N2O5 in China should be acknowledged here.

Wang T. et al., Observations of nitryl chloride and modeling its source and effect on ozone in the planetary boundary layer of southern China, J. Geophys. Res., 121, 2476-2489, 2016.

Wang X. et al., Observations of N2O5 and ClNO2 at a polluted urban surface site in North China: High N2O5 uptake coefficients and low ClNO2 product yields. Atmospheric Environment, 156, 125-134, 2017.

Pg 9, Lines 230-233: it should be noted here that these trends were derived from various observations obtained from different sites in the specific regions, other than

from long-term observations at the same site.

Pg 23, Lines 666-669: cite the final ACP paper instead.

Wen, L. et al., Summertime fine particulate nitrate pollution in the North China Plain: increasing trends, formation mechanisms, and implications for control policy, Atmospheric Chemistry and Physics, 18, 11261-11275, DOI: 10.5194/acp-18-11261-2018, 2018.

Figure 1: the above reference (Wen et al., 2018) has reported very recent observations of fine particulate nitrate at three different sites (urban, rural and mountain sites) in the North China Plain. It would be useful to include these recent data in Figure 1 for comparison.

Figure 4: is the nitrate/sulfate ratio mass-based or molar-based? The molar ratio of nitrate to sulfate should be better here.

Figure 9: please provide a legend for the wind vectors.

Table 1: it should be helpful to provide the exact values of these rate constants used in this study.

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