

Reply on comments

We appreciate the reviewer for the careful reading and their constructive comments on our manuscript. **As detailed below, the reviewer's comments are in normal font, and our responses to the comments are shown in italicized font. New or modified text is in blue.**

All the line numbers refer to the original version of Manuscript ID: **egusphere-2024-3804**

Overall comments:

This work performed direct measurements of N_2O_5 heterogeneous uptake coefficients on atmospheric aerosols in southwestern China and further compared the measured results with those predicted based on different parameterization scenarios. Considering that most of previous studies focusing on gas uptake kinetics are conducted under laboratory conditions, the obtained uptake coefficients may be deviate from the real case in the ambient air. The authors in this work made a step forward and quantified the uptake coefficients of N_2O_5 on real ambient aerosols. The results will help to constrain the uptake coefficients of N_2O_5 to a more realistic basis and further improve our understanding of the heterogeneous reaction kinetics under ambient conditions and their potential impacts on aerosol formation at least locally. Generally, this work could be a meaningful addition into the literature. However, there are several major issues (as pointed out below) need to be addressed at the present stage.

Major issues:

1. Regarding the methods, the related descriptions on the experiment using the aerosol flow tube system are missing key messages. Please give a more detailed description about the air sampling system. Did ambient air directly enter into the flow tube? The measurement system can generate N_2O_5 by itself, but how did you deal with the N_2O_5 in the ambient air? This would also influence the obtained uptake coefficient. For

uptake coefficient calculation, the authors took the flow tube wall effect into account, how did the authors consider the effect caused by aerosol wall losses? Did the wall loss also apply for the gas and particles? What uncertainties would the authors expect for the obtained uptake coefficients?

Thanks for your valuable suggestions. In this aerosol flow tube system, ambient air is directly introduced into the system through a stainless steel sampling tube which removes the ambient N_2O_5 efficiently. The sampling air then mixes with N_2O_5 generated from an N_2O_5 source before entering the aerosol flow tube. After the mixing of ambient air and N_2O_5 source, the total concentration of N_2O_5 at the top of the flow tube is quantified by a cavity-enhanced absorption spectrometer (CEAS-PKU) before the gas enters the flow tube. This, combined with the measured N_2O_5 concentration at the bottom of the flow tube and other parameters, is used to calculate $\gamma(N_2O_5)$. The wall of the flow tube indeed causes the wall loss of aerosols. In previous experiments, we measured the loss of size-resolved Sa at both the top and bottom of the flow tube and determined the loss coefficient to correct for this loss. The total Sa loss caused by the flow tube is approximately 5%. The effect due to wall loss of N_2O_5 gas in the flow tube is mitigated by calibrating k_{wall} every 20 min. We added detailed descriptions and uncertainties of measured gamma via the aerosol flow tube system in Supplementary Information as follows.

“S1. Detailed description of the measurement and calculation of $\gamma(N_2O_5)$

The Aerosol Flow Tube System (AFTS) can be divided into three main modules: the sampling control module, the reaction module, and the detection module. The sampling of the flow tube is facilitated by a vacuum pump located at the end of the system. In the sampling control module, ambient air is directly introduced into the reaction pathway. The sampling gas passes through a 1-in/2-out solenoid valve that directs the sample either through a HEPA filter to remove aerosols or bypasses it, thereby controlling the presence of aerosols in the reaction module. The sampling gas is then mixed with a high concentration of N_2O_5 generated from a N_2O_5 source before entering the reaction module. At the top of the reaction module, two stainless steel static

mixers are installed to ensure that the gas is thoroughly mixed. The aerosol flow tube is the primary site for N_2O_5 uptake reactions.

During detection, concentrations of NO_x and O_3 are continuously measured at the top of the flow tube to facilitate subsequent simulation of gas-phase reactions within the flow tube using a box model. The measurement of N_2O_5 concentration is conducted through two separate 20-minute processes: one to determine the N_2O_5 loss rate in the absence of aerosols (k_{wall}) and another in the presence of aerosols ($k_{\text{wall}}+k_{\text{aerosol}}$). The only difference between the two processes is the presence or absence of aerosols. Each process includes two steps: measuring N_2O_5 concentrations at both the top and bottom of the flow tube, each step maintains 10 min. Throughout the measurement process, the aerosol surface area (Sa) is continuously measured at the bottom of the flow tube, followed by size-resolved Sa correction based on previously determined particle loss coefficients (Chen et al., 2022).

By inputting the measured concentrations of NO_x , O_3 , and N_2O_5 at the top of the flow tube under both aerosol-free and aerosol-present conditions into the box model, the $\text{NO}_3\text{-N}_2\text{O}_5$ chemical reactions and related gas-phase reactions in the flow tube are simulated until the model's output N_2O_5 concentration matches the measured value at the tube's bottom. This process yields k_{wall} and $k_{\text{wall}}+k_{\text{aerosol}}$, from which the N_2O_5 loss rate on aerosols (k_{aerosol}) is derived by subtraction. The $\gamma(\text{N}_2\text{O}_5)$ can then be calculated using established formulas (EqS1).

$$k_{\text{N}_2\text{O}_5} = 0.25 \times \text{Sa} \times \gamma \times C \quad (\text{EqS1})$$

The uncertainty in $\gamma(\text{N}_2\text{O}_5)$ is relevant to the measurement uncertainties of each instrument and the rapid fluctuations of various parameters. To ensure accurate measurements, a rigorous data screening process was implemented. A 10% cutoff for N_2O_5 variation was applied to exclude air masses that were too unstable for valid analysis according to our data screening criteria. Cases showing more than a 2% variation in relative humidity (RH) between HEPA inline and bypass modes were excluded due to RH's significant influence on k_{wall} of N_2O_5 in the aerosol flow tube. To ensure significant N_2O_5 concentration differences due to heterogeneous uptake reactions between the top and bottom of the flow tube, periods with low Sa conditions

($<100 \mu\text{m}^2 \text{ cm}^{-3}$) were filtered out. Additionally, cases where NO concentration exceeded 7 ppbv were excluded to avoid significant changes in $\text{NO}_3\text{-N}_2\text{O}_5$ concentration due to NO titration in the flow tube.

Therefore, the system may introduce a 2% measurement bias in $\gamma(\text{N}_2\text{O}_5)$ due to N_2O_5 concentration fluctuations, a bias of $\pm 8 \times 10^{-4}$ to $\pm 2 \times 10^{-3}$ due to RH fluctuations, a 16% uncertainty from Sa measurement and particle loss in the flow tube, a 4% measurement fluctuation from Monte Carlo simulations, up to a 9% uncertainty from ambient temperature variations, and a 5% uncertainty from NO_x and O_3 concentration fluctuations. In summary, considering all the factors and their corresponding varying ranges discussed above, the overall uncertainty of $\gamma(\text{N}_2\text{O}_5)$ determined from Monte Carlo simulations ranges from 16% to 43%.

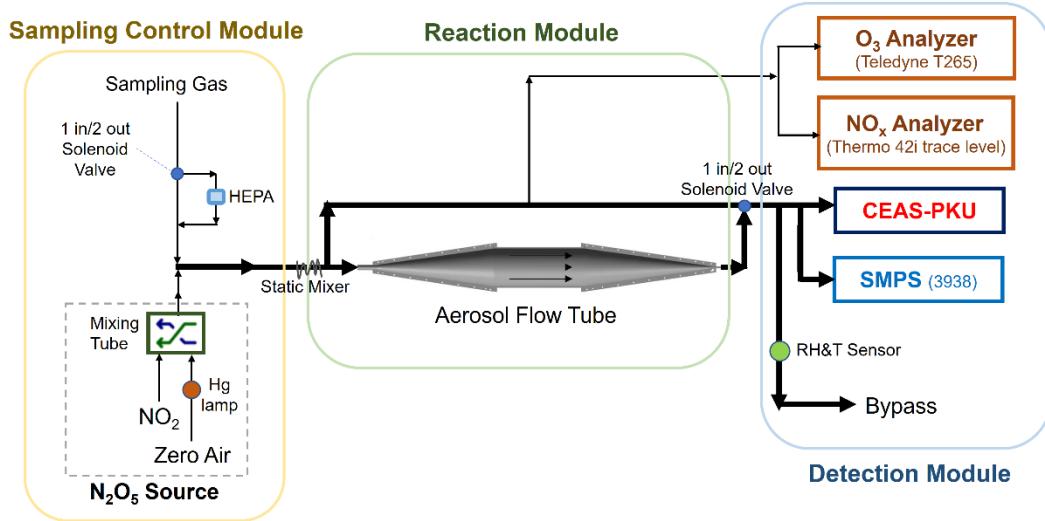


Figure S1. Overall schematic of aerosol flow tube system. Bold arrows indicate the main lines of the sampling gas. “

2. Even though for some cases the parameterized gamma agreed well with the measured gamma (median values), the correlations between them were very bad for all the parameterization scenarios. It would be more helpful to have additional discussions regarding this. Is this more likely caused by parameterization methods or the measurement method? More suggestions on how to choose the different parameterization scenarios under various conditions would be more meaningful from the modelers' point of view.

Thanks for the suggestion. The poor correlation is mainly attributed to the response coefficients of impacting factors in current parameterizations, which failed to reproduce the observations at very high or low levels of these factors. Our analysis identified that inappropriate ALWC response coefficients in current parameterizations contributes to the bias of parameterizations. As shown in Figure 5 of the main text, at low ALWC, all six parameterizations showed overestimation with the maximum difference for EJ05 (189%) and the minimum for MD18 (34%). At median ALWC, the deviation of parameterized $\gamma(N_2O_5)$ reduced to -8~4%. At high ALWC, the parameterizations tend to underestimate the measured $\gamma(N_2O_5)$ with the difference ranging from -37% to -1%. Therefore, we recommend conducting kinetic experiments under extreme ALWC conditions to enhance the fitting efficacy of the parameterizations. A better performance of current parameterizations may also be realized by including parameters, such as particle morphology, phase state, and mixing state (You et al., 2014; Shiraiwa et al., 2017; Ng et al., 2010). These parameters, which are difficult to measure in field studies, have been demonstrated to affect $\gamma(N_2O_5)$. In previous research, McDuffie quantified $\gamma(N_2O_5)$ using a box model and also found poor agreements between the 14 parameterized and $\gamma(N_2O_5)$ values (McDuffie et al., 2018).

We added and revised the discussion in section 3.4 in the main text as follows:

“This phenomenon was possibly caused by several aspects, including the inaccurate estimation of response coefficients of aerosol compositions, relative rates constants of competitive reactions, and the missing parameters. The missing influencing factors in current parameterizations include parameters such as particle morphology, phase state, and mixing state (You et al., 2014; Shiraiwa et al., 2017; Ng et al., 2010). These parameters, which current methodologies are difficult to measure in field conditions, have been proven to affect $\gamma(N_2O_5)$, and can contribute to the discrepancy between parameterized and measured values.”

We added some suggestions on how to choose the different parameterization scenarios under various conditions in section 3.3 in the main text as follows.

“The commonly used parameterizations mainly consist of inorganic and inorganic+organic framework, such as BT09w/oCl, YU20, and MD18. In this study,

among all parameterizations, YU20 demonstrated the best performance, most likely because YU20 was optimized based on datasets observed in four rural regions in China. BT09w/oCl also performed well in this study, overestimating the median by only 7%. However, poor performances of BT09w/oCl were still reported in Pearl River Delta and North China Plain (Wang et al., 2022; Wang et al., 2020). Conversely, the BT09w/oCl performed well in Northwestern Europe, mainly because $\gamma(\text{N}_2\text{O}_5)$ in Europe is predominantly controlled by the ions in bulk phase (Morgan et al., 2015; Chen et al., 2018; Phillips et al., 2016). In North America, $\gamma(\text{N}_2\text{O}_5)$ is significantly inhibited by organic effects (Chang et al., 2016). The parameterizations considering organic effects, like MD18, might be more suitable for the conditions in North America. However, in this study, MD18 showed an overestimation of up to 20%, suggesting that this parameterization is not suitable for China, but more applicable to North American regions.

Hence, most regions in China, where $\gamma(\text{N}_2\text{O}_5)$ is controlled by aerosol liquid water content, are more suited to the YU20. European regions, where gamma is controlled by $\text{H}_2\text{O}/\text{NO}_3^-$ and less influenced by organics, are better served by the BT09w/oCl. Meanwhile, MD18 is more appropriate for North American regions. Localized parameterizations established on the basis of local measurements can exhibit superior performance within the respective regions. Parameterizations incorporating organic effects generally exhibit larger errors than others, underscoring the importance of further improving the consideration of organic effects in parameterizations.”

3. The presentation quality of manuscript seems to be poor. The use of the language appears to be a big problem. Some mistakes should have been avoided if the authors carefully inspect the text before the submission. As shown below, additional edits need to be done regarding the “low level” grammar mistakes. Please note that these grammar issues are not limited to the following list. The authors should therefore check through the whole manuscript very carefully for the revised version.

Minor suggestions/edits:

Line 41-42: What does “response coefficients” mean?

The response coefficients represent the quantitative relationship between $\gamma(N_2O_5)$ and aerosol chemical compositions in parameterizations. Such as k_3 and k_{2b} in BT09 (Eq R1). It corresponds to the fitted relative rates of competing reactions (Yu et al., 2020), or represents functional relationships between aerosol chemical components and $\gamma(N_2O_5)$ (McDuffie et al., 2018).

$$\gamma = \frac{4V_a}{cS_a} K_H k'_{2f} \left(1 - \frac{1}{\left(\frac{k_3[H_2O]}{k_{2b}[NO_3^-]} \right) + 1 + \left(\frac{k_4[Cl^-]}{k_{2b}[NO_3^-]} \right)} \right) \quad Eq\ R1$$

where,

$K_H = 51$, Henry's Law Coefficient (Fried et al., 1994)

$$k'_{2f} = \beta \cdot \beta_e^{(-\delta[H_2O])}$$

$$\beta = 1.15 \times 10^6 \text{ (s}^{-1}\text{)}$$

$$\delta = 0.13 \text{ (M}^{-1}\text{)}$$

$$\frac{k_3}{k_{2b}} = 0.06$$

$$\frac{k_4}{k_{2b}} = 29$$

We have clarified it clearly as follows.

“Our findings suggest the need for more direct field quantifications of $\gamma(N_2O_5)$ and the laboratory measurements under extreme ALWC conditions to re-evaluate the response coefficients between $\gamma(N_2O_5)$ and aerosol chemical compositions in parameterizations.”

And the explanation has been added in section 3.4 as follows.

“This phenomenon was possibly caused by several aspects, including the inaccurate estimation of response coefficients of aerosol compositions (represents the quantitative relationship between $\gamma(N_2O_5)$ and aerosol chemical compositions), relative rates of competitive reactions, and the missing parameters.”

Line 91-95: “However, the comparisons of” Please rephrase this sentence, as it reads unclearly and awkwardly.

It has been revised as follows.

“However, these parameterizations usually exhibit low correlations with observed $\gamma(\text{N}_2\text{O}_5)$ in varying environments (Brown et al., 2009; Ryder et al., 2014; McDuffie et al., 2018).”

Line 96: “can leads to” should be “can lead to”.

Line 105: “European” should be “Europe”.

The above two comments have been revised accordingly in the main text.

Line 113-114: “We further notice …” the whole sentence reads awkwardly. Please rephrase it.

It has been revised as follows.

“We further observe significant biases when estimating particulate nitrate formation potential based on current $\gamma(\text{N}_2\text{O}_5)$ parameterization.”

Line 126: What does CNST mean?

We have clarified it clearly as follows.

“Sunrise was around 06:30 CNST (Chinese National Standard Time = UTC + 8 h) and sunset was at 19:30 CNST.”

Line 129: “Google Maps images” should it be “Google Map images” ?

Line 138: “and” should be “which”.

Line 147: “A total of 117 kinds of VOCs …” could be like “A total of 117 VOCs species …”.

Line 155: “included” should be “including”.

Line 158: For the header of Table 1, “Detection of limit” should be “Detection limits”, “Method” should be “Methods”, “Accuracy” should be “Accuracies”.

The above five comments have been revised accordingly in the main text.

Line 160: “The” is not needed in front of “measurement”, for this case. The same applies to the title of the other sections.

We have revised titles to “2.3 Measurement and calculation of $\gamma(\text{N}_2\text{O}_5)$, 2.4 Calculation of NO_3 and N_2O_5 reactivity, 2.5 Calculation of nitrate production rate, 3.4. Impact of ALWC on parameterized $\gamma(\text{N}_2\text{O}_5)$ ”

Line 165: “(excess)” should be “(in excess)”.

Line 166: “consist of” should be “consists of”.

Line 170: “concentration” should be “concentrations”.

The above three comments have been revised accordingly in the main text.

Line 169-176: These three sentences read awkwardly and apparently have several grammatical mistakes. Please rewrite them.

We rewrite it as follows.

“The detection instruments used for measurements of N_2O_5 , NO_x , O_3 and S_a are CEAS-PKU, Thermo 42i-TL, Teledyne T265 and SMPS (TSI Model 3938). Additionally, a RH&T sensor (Rotronic, Model HC2A-S) was utilized to monitor relative humidity and temperature inside the flow tube. During each duty cycle, N_2O_5 concentrations were recorded both at the inlet and exit of the flow tube under the condition with and without aerosols to derive the wall loss of N_2O_5 . NO , NO_2 , and O_3 concentrations were consistently measured at the inlet of the flow tube, and S_a concentrations were consistently measured at the exit of the flow tube.”

Line 192: “the reaction rate of constant of...” should be “the reaction rate constant of ...”.

We have revised it accordingly in the main text.

Line 209: “The mean diurnal of measured...” should be “The mean diurnal variation of the measured ...”. The authors seem to have a big problem on how to correctly use “the”.

We sincerely appreciate your careful check. We have revised it accordingly and also check this problem in our manuscript thoroughly.

Line 216: “(PNO₃)” should be “P(NO₃)”.

Line 218: “observation campaign” is not the common way. Either “observation period” or just “campaign”.

The above two comments have been revised accordingly in the main text.

Line 252: “Functional dependence” reads very awkwardly. Please change it.

We have revised this title to “3.2. Dependence of γ(N₂O₅) on impacting factors”.

Line 254: What is “organic wet mass fraction”? How did you measure it?

We added the explanation in supporting information as follows.

“S2. Calculate of organic wet mass fraction.

The organic wet mass fraction is defined as the mass fraction of organics within the aerosol containing water. The calculation of organic wet mass fraction is presented as follows.

Organic wet mass fraction=Organic mass/(Organic mass+NO₃⁻ mass+Cl⁻ mass+SO₄²⁻ mass+NH₄⁺ mass+H₂O mass)”

Line 260-262: As the authors stated, “the negative correlation of particulate organic and γ(N₂O₅) was usually weak derived from field measurements”. In the present work, however, the authors found a significant negative correlation between the organic wet mass fraction and the gamma. Could the authors discuss why this study shows such a difference?

Thanks for your valuable comments. We added the discussion in section 3.2 in the main text as follows.

“The organic wet mass fraction in this study varies between 0.3 and 0.8, while other previous studies have reported a variation range of 0.1 to 0.5(McDuffie et al., 2018;Wang et al., 2020;Brown et al., 2009). The large proportion and variation range of organics in the aerosols may lead to a more significant inhibition effect on γ(N₂O₅). Additionally, we found that both the dry and wet mass fractions of organics in this study

showed significant negative correlations with ALWC, with Pearson coefficients of -0.66 and -0.79 (Table S3), respectively. Therefore, organics might decrease $\gamma(\text{N}_2\text{O}_5)$ by forming an organic coating to limit the penetration of liquid water into the particle phase and hinder the reaction between N_2O_5 with liquid phase.”

Line 275: “showed” should be “shows”.

This comment has been revised accordingly in the main text.

Line 290: In the caption of Fig. 3, the authors state that “The points represent the median in each bin,...”. How do the authors select the different bins? Should be the symbols or the points represent the median values?

We basically divided the range of each aerosol chemical component equally into six bins while with some exceptions. The organic dry mass fraction exhibits a discontinuity beyond 0.6 and thus we set the sixth bin at 0.7. Due to the discontinuity in aerosol nitrate concentration changes beyond 3.5 M, an additional bin was added at 4.75 M. Similarly, since mol Cl/mol NO_3^- exhibits a discontinuity beyond 0.4, the sixth bin was positioned at 0.6. We revised the statement of Figure 3 as follows.

“The gray points represent the measured values. The symbols in different colors represent the median in each bin with range from the 10th to 90th percentile in each bin denoted as lines.”

Line 338: “(aka. low ...)”, for me it is the first time to see the use of “aka.”. Probably it is not the official way to use it in a scientific research paper?

This sentence has been revised as follows.

“Although some parameterizations performed relatively well in reproducing the median values of $\gamma(\text{N}_2\text{O}_5)$, none of the ten parameterizations were able to reproduce the range of measured $\gamma(\text{N}_2\text{O}_5)$ values, as indicated by poor correlations and large RMSE.”

Line 339-340: The authors use “response coefficients” several times throughout the manuscript. At least it is not a scientifically meaningful definition, as far as I know. What

is the exact meaning of this? Any references?

Thanks for your comment. We have provided a detailed explanation regarding this issue in the response for Lines 41-42, and we sincerely hope it solves your concern. Response coefficient means the coefficients between $\gamma(N_2O_5)$ and aerosol chemical compositions in parameterizations. Such as k_3 and k_{2b} in BT09 (Eq R1). It corresponds to the relative rates of competing reactions (Yu et al., 2020), or represents relationships between aerosol chemical components and $\gamma(N_2O_5)$ (McDuffie et al., 2018). The explanation has been added in section 3.4 as follows.

“This phenomenon was possibly caused by several aspects, including the inaccurate estimation of response coefficients of aerosol compositions (represents the quantitative relationship between $\gamma(N_2O_5)$ and aerosol chemical compositions), relative rates of competitive reactions, and the missing parameters.”

Line 358-361: Please rephrase the whole sentence to make it more scientifically readable.

This sentence has been revised as follows.

“The overestimation at low ALWC and underestimation at high ALWC suggest that the treatment of coefficients related to ALWC in most parameterizations can hardly capture the response of $\gamma(N_2O_5)$ to largely varied ALWC.”

Line 377-379: Same as above

This sentence has been revised as follows.

“Therefore, we suggest that future studies should conduct more $\gamma(N_2O_5)$ measurements under extreme ALWC levels, which helps to improve the reliability of response coefficients between $\gamma(N_2O_5)$ and ALWC in ambient conditions.”

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