

Reply on RC1

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

All the line numbers refer to original version of Manuscript ID: **amt-2022-167**.

This paper provides a modification to ambient flow tube systems developed by Bertram et al 2009 and Wang et al 2018. The authors document the increased robustness due to measurements prior and after the flow tube as well as a box model to predict side reactions. While this is an interesting finding, I do not think this work is novel enough for publication in AMT. This work belongs as a technical note. I also noted that the authors need to credit Bertram et al 2009 more for use of their design. My comments are below.

We thank this comment. The reviewer emphasized the similarity of our work to the two previous works and questioned the novelty of this study. While our work builds on the previous two studies and attempts to address some issues that were not well addressed in the above work.

First, accurate quantification of wall loss is essential for the quantification of N_2O_5 uptake coefficients as suggested in Bertram's paper, a better way to reduce the wall loss interference is to measure it frequently (Bertram et al., 2009, see section 4.3). This suggestion was subsequently adopted by Wang et al. but the concentration of the N_2O_5 source they used in determining the wall loss and the total N_2O_5 loss was an assumed stable value rather than an observed one. Here, we dynamically determine the N_2O_5 source concentration frequently, which is helpful to provide accurate data on quantifying the wall loss as well as the total N_2O_5 loss in the flow tube in each measurement cycle. Figure R1 shows the example of difference of measured N_2O_5 wall loss at lab condition and ambient condition.

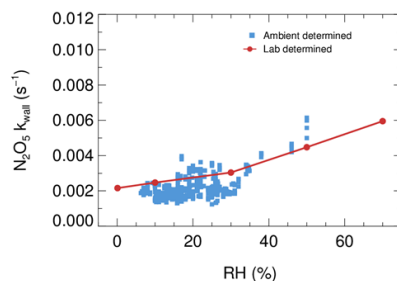


Figure R1. The derived dependence of N_2O_5 wall loss on RH at laboratory condition (red dots) and field measurement (blue square)

Second, the use of an iterative box model approach to correct the potential bias, due to side reactions in the flow tube, is the highlight of the work of Wang et al. The iterative box model calculates backward to estimate the concentration of NO_2 and O_3 before entering the flow tube, with estimated NO profile and measured N_2O_5 at the exit of flow tube. Here, the observed concentrations of NO_2 , O_3 , and NO at “time zero” are obtained through programmed cyclic measurements, which can reduce the uncertainties by adding the model constrain (more details can be found in the reply for the following Question NO. 4). Figure R2(a) presents the box whisker of N_2O_5 and NO concentration before the entrance during a field campaign. The variation of initial N_2O_5 is much larger than that in lab condition with a very small standard deviation for N_2O_5 concentration ($<1\%$, mentioned in line 180), highlights the influence of ambient air mass (e.g. varying NO and temperature) to injected N_2O_5 . Figure 2R(b) shows the case of large underestimation by using a fixed initial N_2O_5 concentration rather than measured values as box model input. In addition, we simulate NO_3 - N_2O_5 relationship via specific reactions rather than approximating it in equilibrium and introducing the equilibrium coefficient (K_{eq}) into calculation. Determining NO_3 or N_2O_5 concentration by K_{eq} could induce large bias (up to 90%) under the high aerosol loading and low temperature (Chen et al., 2021).

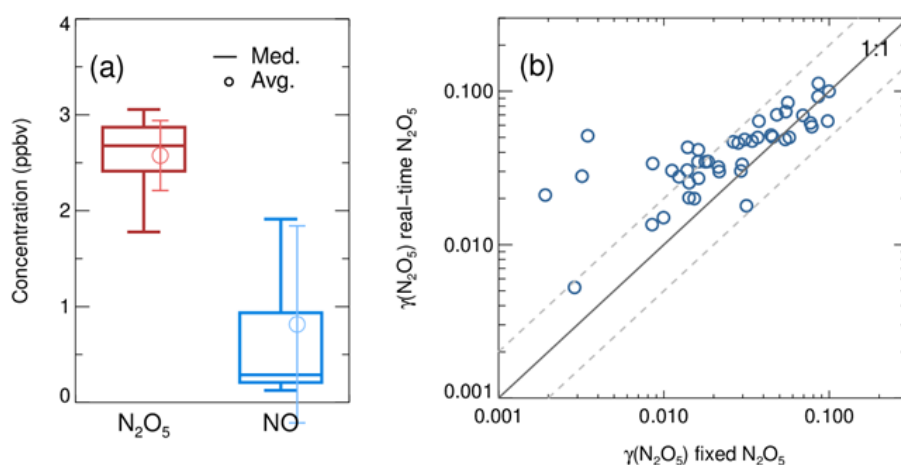


Figure R2. (a) the box whisker of N_2O_5 source and NO measured before the entrance; (b) the inter-comparison of derived N_2O_5 uptake coefficient by using a fixed N_2O_5 and a dynamic measured N_2O_5 at the time zero in the iterative box model.

Third, to achieve the programmed cyclic measurement of key parameters such as N_2O_5 , NO_x and O_3 , we adopted a new design scheme of Y-tee and cyclic measurement setup, which were never used in the previous two studies.

In summary, we believe that the above innovation points of our study advance the field measurement of N_2O_5 uptake coefficient and be enough for publication in AMT, while we may not address our innovation points well in the previous version. Thus, we have added an explanation of the novelty, and credited Bertram et al 2009 and Wang

et al 2018 more for use of their design in the revised manuscript. The detailed revision can be found in the following point to point responses.

MAJOR COMMENTS AND CONCERNS

1. Sections 2.1-2.3, the bulk of the flow tube design, are all pretty much the same as (T. H. . Bertram et al., 2009) yet the authors do not cite this paper in these sections for the design. A reader who has not read Bertram et al, 2009 might very well think that these aspects of the design are the authors’.

We thank the reviewer for pointing out this issue. We have added the citation of this paper and added the statements to clarify the similarity and difference between Bertram et al. (2009) and this work in the beginning of section 2 and section 2.3. The description is modified as follows:

“The design of sampling module and aerosol flow tube in this work follows previous work for measuring $\gamma(\text{N}_2\text{O}_5)$ on ambient aerosols (e.g. Bertram et al., 2009). The major distinctions of this system from previous work are continuous monitor of NO_x and O_3 concentration before the inlet of flow tube (after sampling air mixing with N_2O_5 source) and the sequential measurements of N_2O_5 concentration both at the inlet and the exit of flow tube within a duty cycle.”

“The mechanic design of this flow tube follows that used in Bertram et al. (2009), with different length and diffuser angles particularly designed for our typical flow rate.”

2. The residence time in the flow tube is quoted at 156s, which seems way too short for low values of surface area.

Thank you for pointing out this issue and we totally understand the reviewer’s concern. In fact, we mainly focus on the typical episode days with medium to high aerosol loadings (the surface area concentration larger than $500 \text{ um}^2/\text{cm}^3$) in polluted regions, such as Northern China. As indicated in section 5, under the Sa of $500 \text{ um}^2/\text{cm}^3$, the detection limit of this system is 6.4×10^{-4} , which is well below the previous most of previous ambient $\gamma(\text{N}_2\text{O}_5)$ measurement results ranging from 1×10^{-3} to >0.1 in polluted regions of China (Wang et al., 2020; Wang et al., 2017c; Wang et al., 2017d; Xia et al., 2019). The residence time determined in this work is also slightly higher than 149 s that reported in a previous work focusing on investigating $\gamma(\text{N}_2\text{O}_5)$ in polluted regions (Wang et al., 2018b). In addition, the residence time for our system can be extended to over 300 s by reducing the air flow rate inside the flow tube. The flow rate of current set up is controlled by both the detection instruments and an extra pump with 1 SLPM flow rate attached to the bottom of the flow tube, which was actually designed for adjusting the residence time of the flow tube in order

to match different levels of aerosol loadings. We believe this additional set up can satisfy the requirement of ambient $\gamma(\text{N}_2\text{O}_5)$ measurement and research purpose of N_2O_5 uptake under low surface area concentration. More clarifications have been added in section 4.2 as follows.

“As shown in Section 5, the detection limit of this system is 6.4×10^{-4} with Sa of $500 \text{ um}^2 \text{ cm}^{-3}$, which is well below the previous most of previous ambient $\gamma(\text{N}_2\text{O}_5)$ measurement results ranging from 1×10^{-3} to >0.1 in polluted regions of China (Wang et al., 2020; Wang et al., 2017c; Wang et al., 2017d; Xia et al., 2019). The residence time determined in this work is also slightly higher than 149 s that reported in a previous work focusing on investigating $\gamma(\text{N}_2\text{O}_5)$ in polluted regions (Wang et al., 2018b). In addition, the residence time for this flow tube can be extended to over 300 s to satisfy the $\gamma(\text{N}_2\text{O}_5)$ measurement requirements under low Sa by reducing the flow rate of air passing through, which is controlled by an extra pump.”

3. The authors measure very high values of gamma, much higher than observed for most ambient studies: (T. H. Bertram et al., 2009; Riedel et al., 2012b) and most flow tube work with the exception of dust particles (Mitroo et al., 2019; Tang et al., 2016), I wonder if the authors have underestimated the particle surface area by not using an APS or if the filter upstream of the CEAS is causing an artificially higher gamma than expected.

Thank you for pointing out this issue. The $\gamma(\text{N}_2\text{O}_5)$ measured in this work ranged from 0.0045 to 0.12, which was within the range of 10^{-5} to >0.1 determined from ambient N_2O_5 measurements around the world (Bertram et al., 2009; Brown et al., 2009; McDuffie et al., 2018; Morgan et al., 2015; Tham et al., 2018) and comparable to previous results reported in polluted regions in China (Wang et al., 2020; Wang et al., 2018a; Wang et al., 2017c; Wang et al., 2017d; Xia et al., 2019; Yu et al., 2020). Although it is true that the average of $\gamma(\text{N}_2\text{O}_5)$ (0.042) measured in Beijing 2020 was somewhat higher than other reported average values, this phenomenon is majorly associated with the air mass encountered during measurement period. Owing to the data filtering criterion applied in this study, the valid $\gamma(\text{N}_2\text{O}_5)$ was mostly measured within the air mass came from the North of Beijing with low NO and VOCs concentration (see the following Figure R3). The simultaneous aerosol component measurements by ACSM and elements distribution analysis for single particle by TEM (Transmission electron microscope) indicated that the organics accounted for less than 50% of aerosol dry mass and organic coating was not prevalent on aerosols, respectively, during the periods with valid $\gamma(\text{N}_2\text{O}_5)$ data (will be presented in a following paper and see also the following Figure R4). These evidences may explain the high $\gamma(\text{N}_2\text{O}_5)$ values measured in this study.

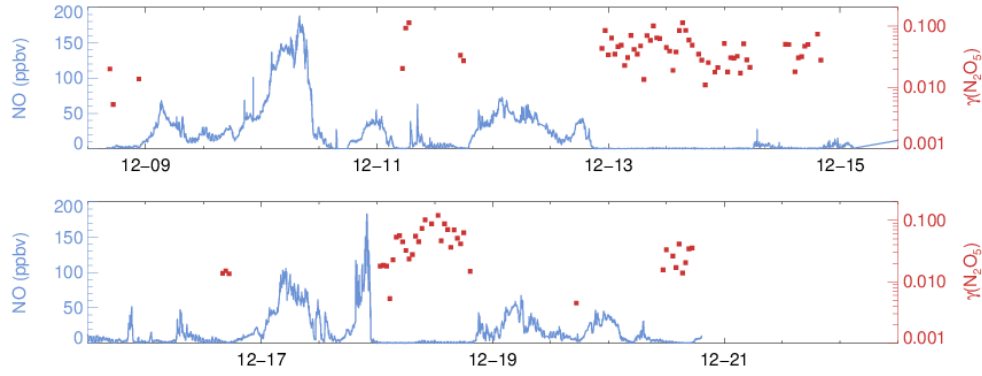


Figure R3. Time series of in-situ measured $\gamma(\text{N}_2\text{O}_5)$ and NO concentration during Dec 09~21 in 2020.

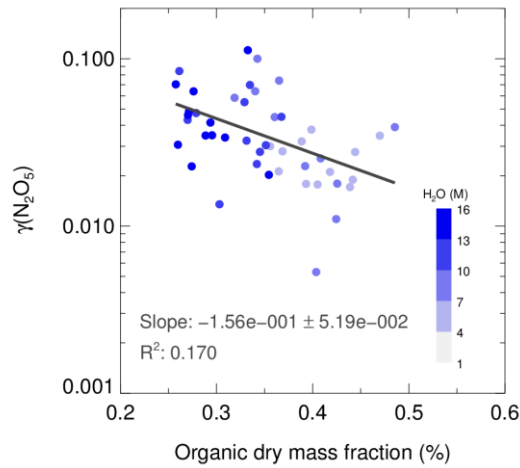


Figure R4. The relation plot of $\gamma(\text{N}_2\text{O}_5)$ and organic dry mass fraction, color coded with aerosol water content.

For the possibility of $\gamma(\text{N}_2\text{O}_5)$ underestimation mentioned by the reviewer, we have included the possible underestimation of S_a and the filter loss of N_2O_5 in the uncertainty analysis. According to on-site characterization of particle size distribution (shown in section 4.1), the S_a concentration distribution peaked at 200-300 nm and the particles larger than 730 nm was estimated to account for less than 5% of S_a concentration. The membrane filter was changed every 2 hours during the measurement period and the transmission efficiency of N_2O_5 on the used membrane was determined to be $92 \pm 3\%$ (also see section 2.4). Such an 8% loss of N_2O_5 was corrected in N_2O_5 measurement. Overall, 5% uncertainty of S_a measurement and 19% uncertainty of N_2O_5 measurement (with 3% resulted from N_2O_5 loss on membrane filter) have been considered in the total uncertainty of $\gamma(\text{N}_2\text{O}_5)$ as indicated in section 5.

4. It is not clear that the box model presented in this work is a significant advance over the box model presented in Bertram et al 2009 and Wang 2018. The authors need to provide evidence that their work is a significant advance over previous work.

We gratefully appreciate for your valuable suggestion. The box model presented in this work can quantify the wall loss rate constant of N_2O_5 and $\gamma(N_2O_5)$, respectively, in one duty cycle. It particularly considers the variation of gas-phase reactions rates and the equilibrium between NO_3 and N_2O_5 , with the constraint of N_2O_5 concentration measured sequentially at both end of the flow tube. In comparison with Bertram et al 2009 and Wang et al 2018, the method we retrieved $\gamma(N_2O_5)$ by using the box model in this work has advances in avoiding the influence resulted from varying air mass (such as varying NO_x , O_3 and RH level), from the equilibrium approximation used in iterative box model and from the variation of initial N_2O_5 concentration.

In the study of Bertram et al 2009, they used the box model to check for the concentration of N_2O_5 source and the uncertainty from gas phase reactions in some cases. Routinely, $\gamma(N_2O_5)$ was retrieved directly by the N_2O_5 concentration measured at the exit of flow tube with and without particles inline. In this work, we used the box model to retrieve $\gamma(N_2O_5)$ based on measurements of NO_x , O_3 and N_2O_5 from time to time. We found that our method can avoid the underestimation of $\gamma(N_2O_5)$ derived by exit-concentration method under different level of NO , NO_2 and O_3 due to the lack of consideration of gas phase reaction. Simulations and laboratory tests on $(NH_4)_2SO_4$ aerosols presented in figure 6 and section 3.2 corroborated that our method can buffer the variation of air mass and the resulted bias from the changes of gas phase reaction rates.

In the study of Wang et al 2018, they used an iterative box model to retrieve $\gamma(N_2O_5)$ based on measurements of NO_x , O_3 and N_2O_5 only at the exit of the flow tube. There are two basic assumptions within the simulation of the iterative box model: The first one is that NO_3 and N_2O_5 are always in equilibrium and equilibrium coefficient Keq can be represented by simple parameterization to derive the NO profile; The second one is that initial N_2O_5 is stable during field measurements. However, in our previous study, the simple parameterization of Keq was found not applicable under high aerosol loading or low temperature, which could lead to over 90% overestimation on Keq value in polluted episode days (Chen et al., 2022). In addition, the initial N_2O_5 concentration after mixing with sampling air is not expected to be as stable as that observed in laboratory tests, due to the variations in temperature, NO concentration and other related parameters. Therefore, the direct constraint of initial N_2O_5 , NO_x and O_3 concentration measured at the inlet of the flow tube in this work enable a straightforward simulation of NO_3 - N_2O_5 chemistry occurring in the flow tube, which reduces the uncertainty of $\gamma(N_2O_5)$ derivation. We have rephrased the statement about the advances of model used in this work and presented it in section 3.

SPECIFIC COMMENTS

Abstract

1. “newly developed” on line 19. From the paper, this just seems like a slight modification instead.

Thanks for your comment. We have rephrased it as follows.

“An improved aerosol flow tube system coupled with detailed box model was developed to measure N₂O₅ heterogeneous uptake coefficients ($\gamma(\text{N}_2\text{O}_5)$) on ambient aerosols directly.”

Introduction

2. Line 47, also cite (Gaston and Thornton, 2016; Mitroo et al., 2019; Riedel et al., 2012a, 2013)

Thanks for your suggestion. We have cited the references as mentioned by the reviewer.

3. Line 59, also cite (Cosman et al., 2008; Escorcia et al., 2010; Folkers et al., 2003; Gaston et al., 2014)

Thanks for your suggestion. We have cited the references as mentioned by the reviewer.

4. Lines 62-65, also mention particle size as well (Gaston and Thornton, 2016). Also missing papers on organic aerosol (Escorcia et al., 2010; Gaston et al., 2014; Griffiths et al., 2009; Thornton et al., 2003)

Thanks for your valuable suggestion. We have added the particle size as well and cited the papers on organic aerosol mentioned by the reviewer.

5. Lines 71-74 reflect the findings in (Thornton et al., 2003), which should be cited here.

Thanks for your careful check and suggestion. We have cited the reference as mentioned by the reviewer.

6. Lines 74-77 reflect the findings in (Mitroo et al., 2019; Royer et al., 2021), which should be cited here.

Thanks for your careful check and suggestion. We have cited the references as mentioned by the reviewer.

7. Line 105, Mitroo et al 2019 did not use an ambient flow tube.

We are sorry for the incorrect citing and have removed Mitroo et al 2019 here.

Methods

1. Sections 2.1-2.3 are really the design of (T. H. Bertram et al., 2009; T. H. Bertram et al., 2009), as such, the authors must use appropriate citations here.

Thanks for your suggestion. We have added the citation as mentioned by the reviewer here and rephrased the description of our design to distinguish from that in Bertram et al 2009. The description is modified as follows:

“The design of sampling module and aerosol flow tube in this work follows previous work for measuring $\gamma(\text{N}_2\text{O}_5)$ on ambient aerosols (e.g. Bertram et al., 2009). The major distinctions of this system from previous work are continuous monitor of NO_x and O_3 concentration before the inlet of flow tube (after sampling air mixing with N_2O_5 source) and the sequential measurements of N_2O_5 concentration both at the inlet and the exit of flow tube within a duty cycle.”

“The mechanic design of this flow tube follows that used in Bertram et al. (2009), with different length and diffuser angles particularly designed for our typical flow rate.”

2. Lines 215-217, wouldn't the use of a filter upstream of the CEAS cause issues where wet, ambient particles would react with N_2O_5 going into the CEAS and cause a higher gamma than one would expect? That might explain the very high values of gamma observed in ambient.

We totally understand the reviewer's concern. The membrane filter was changed every 2 hours during the measurement period and the transmission efficiency of N_2O_5 on the used membrane was determined to be $92 \pm 3\%$ (also see section 2.4). Such an 8% loss of N_2O_5 was corrected in N_2O_5 measurement and 3% uncertainty of N_2O_5 measurement were also included in the uncertainty analysis of $\gamma(\text{N}_2\text{O}_5)$. The use of a filter upstream of the CEAS and the procedures of membrane changing have been successfully applied in many field campaigns to measure ambient N_2O_5 (Brown et al., 2016; Kennedy et al., 2011; Wang et al., 2017a; Wang et al., 2017b; Wang et al., 2018a). We further add the clarifications on the corrections for N_2O_5 possible loss on filter and cited references to support its applicability on N_2O_5 measurements in section 2.4. The modified texts are as follows.

“Laboratory tests have been conducted to quantified the transmission efficiency of N_2O_5 over the membrane ($92 \pm 3\%$), sampling tube of CEAS (99.7%) and the inside of CEAS (93.6%). The use of a filter upstream of the CEAS and the procedures of membrane changing have been successfully applied in many field campaigns to

measure ambient N₂O₅ (Brown et al., 2016; Kennedy et al., 2011; Wang et al., 2017a; Wang et al., 2017b; Wang et al., 2018a). The loss of N₂O₅ on membrane filter, sampling tube and the detection chamber inside the CEAS were corrected according to transmission efficiency and the detection limit of N₂O₅ was determined to be 2.7 pptv (1 σ , 60s) with the measurement uncertainty of 19%.”

For the high gamma measured in this work, this phenomenon is majorly associated with the air mass encountered during measurement period. Please see also our response to comment 3 in major comment.

3. Lines 287-288, this duty cycle is not that different from (T. H. . Bertram et al., 2009)

Thanks for your suggestion. We have rephrased the description of duty cycle in this work and cited the work by Bertram et al 2009. The modified texts are as follows.

“As a result, a typical duration of duty cycle is composed of 40 minutes with 20 minutes for each mode, which is similar to that in Bertram et al. (2009).”

4. Lines 305-308, what fraction of VOCs measured had known rate constants that can be used to parameterize the reaction of NO₃ with VOCs?

A total of 59 kinds of VOCs were measured by GC-FID-MS in this work, half of which had known rate constants that can be used to parameterize the reaction of NO₃ with VOCs (mainly compose of alkenes and aromatics). Their rate constants were obtained from MCM331 or IUPAC and the values at 298K are listed as follows. We also added clarification about the measured VOCs with known rate constants in section 3.1 and the following table in the appendix.

Table A1. VOCs used to calculate NO₃ reactivity (k_{NO_3}) in the box model method

Species	$k_{\text{NO}_3}(298 \text{ K})$	Species	$k_{\text{NO}_3}(298 \text{ K})$
METHANE	1D-18 ^b	TRANS-2-PENTENE	3.70D-13 ^a
ETHANE	1D-17 ^b	1-HEXENE	1.20D-14 ^a
PROPANE	7D-17 ^b	1-3 BUTADIENE	1.03D-13 ^a
N-BUTANE	4.6D-17 ^b	ISOPRENE	7.0D-13 ^b
I-BUTANE	1.1D-16 ^b	STYRENE	1.50D-12 ^a
ETHYLENE	2.1D-16 ^b	ETHYNE	1D-16 ^b
PROPYLENE	9.5D-15 ^b	BENZENE	3D-17 ^b
1-BUTENE	1.3D-14 ^b	TOLUENE	7.8D-17 ^b
CIS-2-BUTENE	3.50D-13 ^a	O-XYLENE	4.10D-16 ^a
TRANS-2-BUTENE	3.90D-13 ^a	M-XYLENE	2.60D-16 ^a

I-BUTENE	3.4D-13 ^b	P-XYLENE	5.00D-16 ^a
1-PENTENE	1.20D-14 ^a	ETHYL BENZENE	1.20D-16 ^a
CIS-2-PENTENE	3.70D-13 ^a	N-PROPYL BENZENE	1.40D-16 ^a

Note: a. MCM; b. IUPAC

“The reaction of VOCs and NO₃ is treated as pseudo-first-order with a rate constant of $k_{\text{NO}_3\text{-VOCs}}$, which is the sum of rate constants for reactions of NO₃ with each VOCs scaled by the concentration of VOCs measured by GC-FID. In this work, there are 30 kinds of measured VOCs having known reaction rate constants with NO₃ included in the model.”

5. Section 3.2, it’s not clear how this box model differs from the previous studies cited.

Thanks for your comment. The model we used to retrieve $\gamma(\text{N}_2\text{O}_5)$ in this work was directly constrained by N_2O_5 , NO_x and O_3 concentration at the inlet of the flow tube, as well as N_2O_5 and Sa at the exit of the flow tube. In this method, the uncertainty of $\gamma(\text{N}_2\text{O}_5)$ derivation can be reduced owing to avoiding the influence resulted from varying air mass (such as varying NO_x , O_3 and RH level), from the equilibrium approximation used in iterative box model and from the variation of initial N_2O_5 concentration. Please see also our response to comment 4 in major comments for more details as well.

6. Lines 437-441 is similar to the findings of (T. H. Bertram et al., 2009)

Cited the Bertram et al 2009 as follows.

“The results we obtained from above particle transmission experiments are similar to the findings of Bertram et al. (2009).”

7. Lines 489-490, 156 s for a residence time is quite short, especially for low surface areas. What is the time required for complete mixing of N_2O_5 in the flow tube?

Thanks for pointing out this issue. For the question of short residence time, please see also our response to comment 2 in major comments. For the question of the time required for complete mixing of N_2O_5 in the flow tube, we conducted experiments sampling $(\text{NH}_4)_2\text{SO}_4$ aerosols continuously. The measurement of N_2O_5 and Sa at the exit of the flow tube show that it took about 15 minutes for N_2O_5 and Sa to completely mix in the flow tube (as shown in the following figure). The residence time distribution (RTD) profiles (see in section 4.2) also demonstrated that a pulse injection of NO_2 requires 10~15 minutes to be fully released from the flow tube, which to some extent supports the 15-minute time required for complete mixing of N_2O_5 . We have reorganized the statements on these results and presented it in section 2.5 as follows.

“We measured Sa and N₂O₅ concentration continuously at the exit of flow tube when sampling (NH₄)₂SO₄ aerosols. As shown in the following figure, it took about 15 minutes for particles to rise to a stable level from none or to decrease from a certain level to none, when our system underwent mode switches. The correspondingly periodical variation of N₂O₅ concentration was consistent with particles. The residence time distribution (RTD) profiles (see in section 4.2) also demonstrated that a pulse injection of NO₂ requires 10~15 minutes to be fully released from the flow tube, which to some extent supports the 15-minute time required for complete mixing of N₂O₅.”

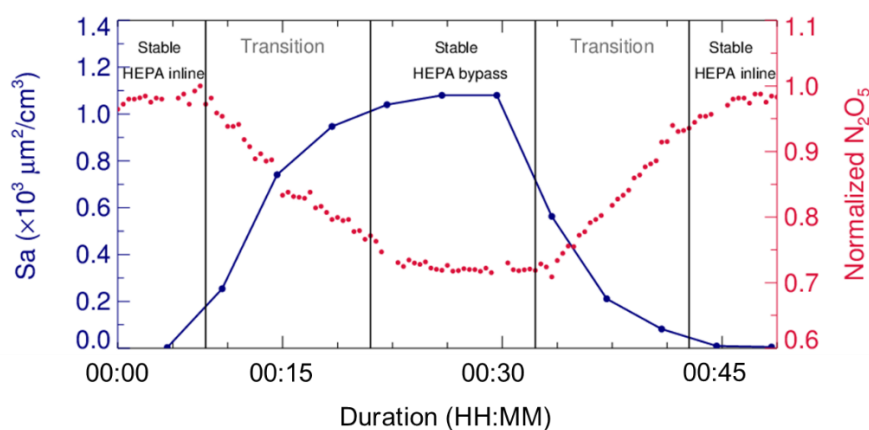


Figure 3. Variations of Sa and N₂O₅ concentration (normalized to peak values) measured at the exit of flow tube when switching the sampling mode. The phases of species concentrations in the flow tube approaching stable after a mode switch are denoted as the transition phases.

8. Lines 635-639, gamma values seem really high. The authors should provide some explanation of how gamma varied as a function of air mass encountered.

Thanks for pointing out this issue and your valuable suggestion. Please see also our response to comment 3 in major comments.

References:

- Bertram, T. H., Thornton, J. A., Riedel, T. P., Middlebrook, A. M., Bahreini, R., Bates, T. S., Quinn, P. K., and Coffman, D. J.: Direct observations of N₂O₅ reactivity on ambient aerosol particles, *Geophys. Res. Lett.*, 36, 10.1029/2009gl040248, 2009.
- Brown, S. S., Dube, W. P., Fuchs, H., Ryerson, T. B., Wollny, A. G., Brock, C. A., Bahreini, R., Middlebrook, A. M., Neuman, J. A., Atlas, E., Roberts, J. M., Osthoff, H. D., Trainer, M., Fehsenfeld, F. C., and Ravishankara, A. R.: Reactive uptake coefficients for N₂O₅ determined from aircraft measurements during the Second Texas Air Quality Study: Comparison to current model parameterizations, *J. Geophys. Res.- Atmos.*, 114, D00F10(01-16), Artn D00f10 10.1029/2008jd011679, 2009.
- Brown, S. S., Dubé, W. P., Tham, Y. J., Zha, Q., Xue, L., Poon, S., Wang, Z., Blake, D. R., Tsui, W., Parrish, D. D., and Wang, T.: Nighttime chemistry at a high altitude site above Hong Kong, *J. Geophys. Res.: Atmos.*, 121, 2457-2475, 10.1002/2015jd024566, 2016.
- Chen, X. R., Wang, H. C., and Lu, K. D.: Interpretation of NO₃-N₂O₅ observation via steady state in high-aerosol air mass: the impact of equilibrium coefficient in ambient conditions, *Atmos. Chem. Phys.*, 22, 3525-3533, 10.5194/acp-22-3525-2022, 2022.
- Kennedy, O. J., Ouyang, B., Langridge, J. M., Daniels, M. J. S., Bauguitte, S., Freshwater, R., McLeod, M. W., Ironmonger, C., Sendall, J., Norris, O., Nightingale, R., Ball, S. M., and Jones, R. L.: An aircraft based three channel broadband cavity enhanced absorption spectrometer for simultaneous measurements of NO₃, N₂O₅ and NO₂, *Atmospheric Measurement Techniques*, 4, 1759-1776, 10.5194/amt-4-1759-2011, 2011.
- McDuffie, E. E., Fibiger, D. L., Dubé, W. P., Lopez-Hilfiker, F., Lee, B. H., Thornton, J. A., Shah, V., Jaeglé, L., Guo, H., Weber, R. J., Michael Reeves, J., Weinheimer, A. J., Schroder, J. C., Campuzano-Jost, P., Jimenez, J. L., Dibb, J. E., Veres, P., Ebben, C., Sparks, T. L., Wooldridge, P. J., Cohen, R. C., Hornbrook, R. S., Apel, E. C., Campos, T., Hall, S. R., Ullmann, K., and Brown, S. S.: Heterogeneous N₂O₅ Uptake During Winter: Aircraft Measurements During the 2015 WINTER Campaign and Critical Evaluation of Current Parameterizations, *J. Geophys. Res.: Atmos.*, 123, 4345-4372, 10.1002/2018jd028336, 2018.
- Morgan, W. T., Ouyang, B., Allan, J. D., Aruffo, E., Di Carlo, P., Kennedy, O. J., Lowe, D., Flynn, M. J., Rosenberg, P. D., Williams, P. I., Jones, R., McFiggans, G. B., and Coe, H.: Influence of aerosol chemical composition on N₂O₅ uptake: airborne regional measurements in northwestern Europe, *Atmos. Chem. Phys.*, 15, 973-990, 10.5194/acp-15-973-2015, 2015.
- Tham, Y. J., Wang, Z., Li, Q. Y., Wang, W. H., Wang, X. F., Lu, K. D., Ma, N., Yan, C., Kecorius, S., Wiedensohler, A., Zhang, Y. H., and Wang, T.: Heterogeneous N₂O₅ uptake coefficient and production yield of ClNO₂ in polluted northern China: roles of aerosol water content and chemical composition, *Atmos. Chem. Phys.*, 18, 13155-13171, 10.5194/acp-18-13155-2018, 2018.
- Wang, H., Chen, J., and Lu, K.: Development of a portable cavity-enhanced absorption spectrometer for the measurement of ambient NO₃ and N₂O₅: experimental setup, lab characterizations, and field applications in a polluted urban environment, *Atmospheric Measurement Techniques*, 10, 1465-1479, 10.5194/amt-10-1465-2017, 2017a.
- Wang, H., Lu, K., Chen, X., Zhu, Q., Chen, Q., Guo, S., Jiang, M., Li, X., Shang, D., Tan, Z., Wu, Y., Wu, Z., Zou, Q., Zheng, Y., Zeng, L., Zhu, T., Hu, M., and Zhang, Y.: High N₂O₅ Concentrations

Observed in Urban Beijing: Implications of a Large Nitrate Formation Pathway, *Environ Sci Tech Lett*, 4, 416-420, 10.1021/acs.estlett.7b00341, 2017b.

Wang, H., Chen, X., Lu, K., Tan, Z., Ma, X., Wu, Z., Li, X., Liu, Y., Shang, D., Wu, Y., Zeng, L., Hu, M., Schmitt, S., Kiendler-Scharr, A., Wahner, A., and Zhang, Y.: Wintertime N₂O₅ uptake coefficients over the North China Plain, *Science Bulletin*, 65, 765-774, <https://doi.org/10.1016/j.scib.2020.02.006>, 2020.

Wang, H. C., Lu, K. D., Guo, S., Wu, Z. J., Shang, D. J., Tan, Z. F., Wang, Y. J., Le Breton, M., Lou, S. R., Tang, M. J., Wu, Y. S., Zhu, W. F., Zheng, J., Zeng, L. M., Hallquist, M., Hu, M., and Zhang, Y. H.: Efficient N₂O₅ uptake and NO₃ oxidation in the outflow of urban Beijing, *Atmos. Chem. Phys.*, 18, 9705-9721, 10.5194/acp-18-9705-2018, 2018a.

Wang, W., Wang, Z., Yu, C., Xia, M., Peng, X., Zhou, Y., Yue, D., Ou, Y., and Wang, T.: An in situ flow tube system for direct measurement of N₂O₅ heterogeneous uptake coefficients in polluted environments, *Atmospheric Measurement Techniques*, 11, 5643-5655, 10.5194/amt-11-5643-2018, 2018b.

Wang, X., Wang, H., Xue, L., Wang, T., Wang, L., Gu, R., Wang, W., Tham, Y. J., Wang, Z., Yang, L., Chen, J., and Wang, W.: Observations of N₂O₅ and ClNO₂ at a polluted urban surface site in North China: High N₂O₅ uptake coefficients and low ClNO₂ product yields, *Atmos. Environ.*, 156, 125-134, 10.1016/j.atmosenv.2017.02.035, 2017c.

Wang, Z., Wang, W., Tham, Y. J., Li, Q., Wang, H., Wen, L., Wang, X., and Wang, T.: Fast heterogeneous N₂O₅ uptake and ClNO₂ production in power plant and industrial plumes observed in the nocturnal residual layer over the North China Plain, *Atmos. Chem. Phys.*, 17, 12361-12378, 10.5194/acp-17-12361-2017, 2017d.

Xia, M., Wang, W., Wang, Z., Gao, J., Li, H., Liang, Y., Yu, C., Zhang, Y., Wang, P., Zhang, Y., Bi, F., Cheng, X., and Tao, W.: Heterogeneous Uptake of N₂O₅ in Sand Dust and Urban Aerosols Observed during the Dry Season in Beijing, *Atmosphere*, 10, 204, 10.3390/atmos10040204, 2019.

Yu, C., Wang, Z., Xia, M., Fu, X., Wang, W., Yee Jun, T., Chen, T., Zheng, P., Li, H., Shan, Y., Wang, X., Xue, L., Zhou, Y., Yue, D., Ou, Y., Gao, J., Lu, K., Brown, S., Zhang, Y., and Tao, W.: Heterogeneous N₂O₅ reactions on atmospheric aerosols at four Chinese sites: improving model representation of uptake parameters, *Atmos. Chem. Phys.*, 20, 4367-4378, 10.5194/acp-20-4367-2020, 2020.