

The comments from referee #1 are in blue, and our replies are in black. The major changes to the manuscripts are highlighted in red in both the manuscript and this reply.

The research carried out on the uptake of N₂O₅ on TiO₂ was done very systematically and the manuscript is well written. The manuscript describes the determination of the uptake coefficient of N₂O₅ on TiO₂ particles at room temperature for the first time. Since the refractive index of TiO₂ is more than 60% greater than that of H₂SO₄ particles, main light scattering particles in the stratosphere, it requires much less amount to inject into the stratosphere to reduce the global warming. Unlike H₂SO₄, TiO₂ would not presumably activate chlorine production to cause ozone-destructing chain reaction. Consequently, it would increase stratospheric ozone, thereby lowering photolysis rates in the troposphere and increases in N₂O₅ concentration.

I have only some minor comments:

Reply: We appreciate the very positive comments from referee #1, and would like to thank him/her for the recommendation of publishing our manuscript.

1. page 4424, para 1: How much TiO₂ has to be injected into the stratosphere to have a perceptible impact?

Reply: The referee has raised a good question, though it is not easy to define “a perceptible effect”. The use of TiO₂ for SRM requires a factor of ~3 less in mass compared to sulfate. Since in this manuscript we always use the eruption of Mt. Pinatubo as a reference, at the end of this paragraph we have added a sentence in order to give an idea of the amount of TiO₂ needed: “i.e., only 10 Tg TiO₂ particles are needed (Pope et al., 2012)”.

2. What are other pathways for N₂O₅ loss on TiO₂ than just hydrolysis? Is it possible to have NO₂ produced as a result of the uptake? In such a situation, what would be the impact in terms of ozone depletion?

Reply: We believe that the uptake of N_2O_5 onto TiO_2 only leads to the formation of nitric acid and nitrate on the particles, according to previous studies on the heterogeneous reaction of N_2O_5 with mineral particles. To make it clearer, we have added one sentence to the last paragraph in page 4425: “Seisel *et al.* (2005) observed the formation of nitrate on mineral dust particles due to the uptake of N_2O_5 using diffuse reflectance FTIR, and Tang *et al.* (2012) further confirmed that the yield of nitrate is ~ 2 (as expected from R1) within the experimental uncertainty, and that the formation of NO_2 is negligible”.

3. page 4430, lines 15-24: This assumption is fine on a relative scale. However, one N_2O_5 does not give one NO_2 and one NO_3 . There is always some loss of NO_3 to give $\text{NO}_2 + \text{O}_2$.

Reply: We do not quite agree with the referee. In fact this is a well-established method to measure N_2O_5 . At the end of this paragraph we have added a sentence to explain it: “This scheme has been suggested as an absolute method to calibrate other N_2O_5 detection methods (e.g. CIMS) (Fahey *et al.*, 1985), and is widely used to study the heterogeneous reactions of N_2O_5 with aerosol particles (e.g., Wagner *et al.*, 2008).”

4. Page 4434, line 10- 25: It is good to see a detailed and rigorous of the diffusion correction. However, diffusion correction for small uptake coefficient values is negligible.

Reply: As the referee points out, the diffusion correction is negligible because of the small uptake coefficients determined in our study. We prefer to keep the detailed description of the rigorous correction method, because the new aerosol flow tube in the Cambridge lab is described here for the first time here.

Page 4438, line 13: “P25” should be “P2.5”

Reply: This is not a typo. It is the type of TiO_2 , which we used for our study of “P25”.

This paper is by no means a complete study as pointed out by authors regarding the photocatalytic activity of TiO₂. However, it did a comprehensive experiment and discussion of the results on the uptake of N₂O₅ on TiO₂ particles.

Reply: We appreciate the very positive comments from the referee. Indeed as he/she says, this is not a complete study regarding the potential chemistry of TiO₂ particles under stratospheric conditions. We have added two sentences in line 17 of page 4441 to the discussion, highlighting the importance of heterogeneous chlorine activation: “Heterogeneous chlorine activation is not included in the modeling work because of the lack of reliable kinetics data. The uptake of ClONO₂ onto airborne mineral particles is under investigation in an ongoing study, and new laboratory data will be included in the model to assess the effect of heterogeneous chlorine activation on stratospheric O₃ in future work”.

Page 4441, line 29 (last line): “feebaks” should be “feedbacks”.

Reply: Thank you, the typo was corrected.

This manuscript should be accepted after addressing a few minor points.

Reply: We would like to thank the referee for his/her comments and support of publishing our manuscript.

References:

- Fahey, D. W., Eubank, C. S., Hubler, G., and Fehsenfeld, F. C.: A Calibrated Source of N₂O₅, Atmos. Environ., 19, 1883-1890, 1985
- Seisel, S., Borensen, C., Vogt, R., and Zellner, R.: Kinetics and mechanism of the uptake of N₂O₅ on mineral dust at 298 K, Atmos. Chem. Phys., 5, 3423-3432, 2005
- Tang, M. J., Thieser, J., Schuster, G., and Crowley, J. N.: Kinetics and mechanism of the heterogeneous reaction of N₂O₅ with mineral dust particles, Phys. Chem. Chem. Phys., 14, 8551-8561, 2012

Wagner, C., Hanisch, F., Holmes, N., de Coninck, H., Schuster, G., and Crowley, J. N.: The interaction of N₂O₅ with mineral dust: aerosol flow tube and Knudsen reactor studies, *Atmos. Chem. Phys.*, 8, 91-109, 2008.