Response to Anonymous Referee #2

This study developed an improved observation-based parameterization of N2O5 uptake coefficient and showed that the new parameterization improved the simulation results of NO2 and NO3- by the WRF-CMAQ model. The manuscript is generally well written. I think that it can be considered for publication after the authors address the following comments and suggestions.

Response: We appreciate the reviewer for the helpful comments on our manuscript. We have made all of the suggested changes and clarifications. The reviewer's comments are in black and our responses are in blue, and the changes in the manuscript are in red.

1. Please clearly indicate the scope of application of the new parameterization. Is it applicable to China only or the whole world? I suggest that you apply the parameterization to all the sites shown in Fig. 3 to examine how it performs in other regions of the world. Even within China, please comment on whether the five sites used in this study are representative of China's general environmental conditions.

Response: We think the empirical parameterization should be applicable to different areas in China, especially those polluted regions. The four sites were all located in semi-rural areas with regional representativeness in north or south China. For the sites other than described in this study in Fig. 3, the detailed experimental data such as inorganic compositions and V_a/S_a of each data points are not available in the literature, and thus it is not possible for us to evaluate and compare the new empirical parameters at all other sites in the world. We advocate further validation of the parameterization derived from the present study in other regions of the world.

We added more information to clarify it, as follows,

"All the sites are regionally representative sites, as they are situated in an area with limited anthropogenic influences (Tham et al., 2016; Wang et al., 2017a; Yun et al., 2018; Wang et al., 2016). The detailed information of the sampling sites, instrumentation and γ_{N2O5} determination approach have been described in the previous publications (Wang et al., 2016; Tham et al., 2016; Wang et al., 2017a), and site descriptions are briefly summarized in the SI. The locations of all the measurement sites are shown on the map in Fig. 1a. The statistics of the trace gases and $PM_{2.5}$ measured during the campaigns were summarized in Fig. 1b, representing general pollution conditions at these sites. The mean concentration of O_3, NOx and $PM_{2.5}$ at these sites ranged from 43 to 80 ppbv, 2.4 to 14.5 ppbv and 9.9 to 80.2 μg m^{-3}, respectively."

"More tests of this empirical parameterization are warranted for other locations/seasons in China and other parts of the world."

References

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2. You only evaluated the CMAQ simulation results against NO2 and NO3- observations. Since you made many N2O5 and ClNO2 measurements in this study, I strongly suggest that you also compare the simulation results with these data to better evaluate the performance of the new parameterization in CMAQ. In fact, I think the evaluation results of N2O5 may more directly reflect the performance of the N2O5 uptake parameterization.

Response: We agree with the reviewer that the comparison of simulated N_2O_5 could provide a more direct evaluation of the performance of the new parameterization. Because of the short lifetime of N_2O_5 (usually several to ten minutes) (Tham et al., 2018; Yun et al., 2019), it is more prone to be affected by local emissions and fluctuation of meteorological parameters. For the regional models with a grid resolution of tens of kilometers, it is difficult for the regional model to capture the variation of N_2O_5. We made a comparison of the statistic results of simulated N_2O_5 concentrations in winter 2017 with those observed in the wintertime at various locations of China, including two in the North China Plain (Beijing and Wangdu in Hebei province) and two in southern China (Tai Mao Shan and Heshan). With the new parameterization, the WRF-Chem model can better simulate the average concentrations and variation ranges of N_2O_5 at these locations. The results were added as the new Figure 6, and the discussions were also added, as follows,

"In addition to NO_2 and NO_3^-, we also compared the simulated N_2O_5 concentrations for December 2017 with those observed in the wintertime at various locations of China, including two in the North China Plain (Beijing and Wangdu in Herbei province) and two in southern China (Tai Mao Shan and Heshan). As shown in Figure 6, with the new parameterization, the WRF-CMAQ model can better simulate the average concentration and variation range of N_2O_5 at these locations. Overall, the new parameterization has significantly reduced the discrepancies between the modelled and observed concentrations of NO_2, N_2O_5 and NO_3^- at our study sites and periods in both northern and southern China. More tests of this empirical parameterization are warranted for other locations/seasons in China and other parts of the world."

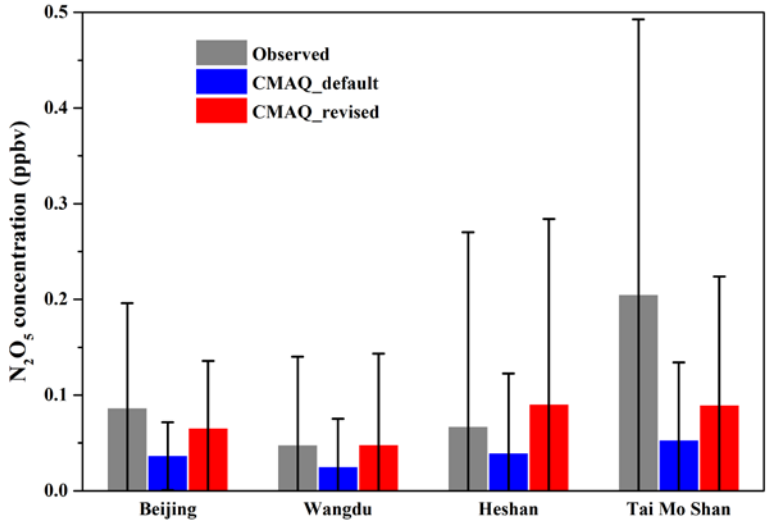

Figure 6. Comparison of the simulated N_2O_5 concentrations by the CMAQ model for December 2017 with the wintertime observation results from four sites in China. The field observations were conducted in December 2017 at Wangdu, January 2018 at Beijing, January 2018 at Heshan and November 2013 at Tai Mo Shan. The columns and error bars represent the average value and standard deviation, respectively.

3. Line 246-249: Your modeling domain covers the whole China and this sentence implies that you do have observational data in southern China. In this case, it looks strange that you only evaluated the simulation results over the North China Plain. I suggest that you provide a quantitative evaluation in southern China rather than just a speculation here.

Response: We focused on northern China for Dec 2017 in evaluating the new parameterization in part because of the availability of the unique regional observations of $PM_{2.5}$ nitrate aerosol. We agree that comparisons with N_2O_5 observations are valuable. As responded to comment #2, we have further compared the model simulation in southern China with our previous field observations at Heshan and Tai Mo Shan, and the comparison results of N_2O_5 are now included in the revised text, see the response to above comment #2.

4. Line 130-132: Although the detailed configuration of CMAQ has been described in a previous paper, I think it is still helpful to briefly describe some key configurations, especially those related to NO2/N2O5/ClNO2/NO3- chemistry.

Response: Adopted and a brief description of CMAQ configuration is added.

"In addition, the Community Multiscale Air Quality (CMAQ) model (v5.1) was employed to evaluate the uptake parameterization. Two simulations (default and revised) were conducted. In the default case, the N_2O_5 uptake and $ClNO_2$ production were calculated based on the parameterization of Bertram and Thornton (2009). In the revised case, the new parameterization derived in this study was used. Other model configurations were the same. The SAPRC07tic

gas mechanism and AERO6i aerosol mechanism was used. Weather Research and Forecasting (WRF) (v4.0) was applied to generate the meteorological inputs for the CMAQ simulations. The anthropogenic emission inputs were generated based on the local Chinese emission inventory (Zhao et al. 2018) and the INTEX-B dataset for Asia (Zhang et al., 2009). The high-resolution chloride emission inventory for China from Fu et al. (2018) was also included. More details for model configuration can be found in Fu et al. (2019). The simulation domain covers China with a resolution of 36×36 km (Fig. S1), based on a Lambert projection with two true latitudes of 25°N and 40°N."

References

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5. Fig. 2d: Obviously the curve does not fit the data points well. Could you justify why you select this formula?

Response: Thanks for pointing this out. The curve was fitted just to check whether the trend following the relationship between γ_{N2O5} and Cl^-/NO_3^- derived from laboratory studies. The discrepancy of the data and the curve shows the Cl^- enhancement in our study is not as strong or obvious as that found in other laboratory studies. To avoid misleading the reader, the fitting curve is removed from Fig. 2d.