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Geoscientific Model Development Discussions

Interactive comment on "Effects of heterogeneous reactions on global tropospheric chemistry" *by* Phuc T. M. Ha et al.

Anonymous Referee #1

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Comments to Ha et al.

This paper evaluated the effects of heterogeneous uptake reactions of N2O5, HO2 and RO2 on cloud and aerosol particles by using a chemical-climate model CHASER, and the modelling results have been verified by comparing with ground-based measurements, shipboard, aircraft and satellite observations. Although the findings of this study on the changes in global abundances of NO2, NO3, O3, and CO, and lifetime of CH4 are basically within the range of uncertainties of previous studies, and no new surprising finding are reported, this work provides the most comprehensive view among this kind of studies covering the lower to upper troposphere, polluted terrestrial and remote oceanic region, and seasonal to annual characteristics. Particularly the study demonstrated the heterogeneous effect in the remote areas such as oceanic region

and the upper troposphere for the first time. Tthe present reviewer judges this paper is acceptable for publication after considering the following comments.

1. The difference between the role of uptake of HO2 and RO2 should be explained more in detail. In the case of the uptake of RO2, the reduction of the formation of PAN and organic nitrates due to the reactions, CH3COO2 + NO2 ïĆő PAN, and RO2 + NO ïĆő RONO2, as well as the reduction of NO oxidation reaction, RO2 + NO ïĆő RO + NO2, RO + O2 ïĆő HO2 etc. are expected. How the difference in the effect of HR(HO2) and HR(RO2) shown in Figs. 11 and 12 can be explained by these factors? 2. Other than the well-known heterogenous processed of N2O5, HO2 and RO2 analyzed in this study, the heterogeneous renoxification process of HNO3 to reproduce NOx has previously been suggested in order to explain the model overestimate of HNO3/NOx ratio in the free and polluted atmosphere (Hauglustaine et al., Geophys. Res. Lett., 23, 2609-2612, 1996: Lary et al., J. Geophys. Res., 102, 3671–3682, 1997; Li et al., SOLA, 11, 124–128, 2015; Akimoto et al., Atmos. Chem. Phys., 19, 603-615, 2019). Although the importance of this process has not been established, the same tendency of overestimate of HNO3 and underestimate of NOx has been revealed in this study (Table 5). Discussion should be given for the possibility of the heterogeneous reaction of HNO3 whether in supporting or objecting. 3. Many of the figures are rather poorly presented for readers and should be revised. (1) In most of the figures, size of inside letters and axis labels are too small (unreadable on print and difficult to read even on PC screen). (2) Fig.3: How the site for each species were selected? There is no explanation in the text. (3) Figs. 3, 4, 5: The difference between the plots for noHR_n2o5, _ho2, _ro2 and _CLD are almost undiscernible. It is suggested to show only noHR and STD in these Figures, and the difference of noHR_n2o5, _ho2, _ro2 and _CLD should be presented in some selected plots in a different Figure. (4) Figs. 9, 11, 12: The differences between the upper and lower figures are not discernible easily. It is suggested to delete the figures for HRS(N2O5-aerosols), HRS(HO2-Cloud) and HRS(RO2-Cloud) in these Figures. It would be enough to explain in the text that the uptake of N2O5 on aerosols, and that of HO2 and RO2 on cloud are major processes.

Explanation should be given in the text why the process predominate for each of the species. (5) Figs. 10, 14: Labels and units of horizontal axis should be given properly. 4. Table 2: What is the meaning of asterisk for “product*”. What do the ISO2 and MACRO2 stand for? 5. Tables 5, 6, 7, 8: Units should be given appropriately.

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