

Geosci. Model Dev. Discuss., referee comment RC1
<https://doi.org/10.5194/gmd-2021-385-RC1>, 2022
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Comment on gmd-2021-385

Anonymous Referee #1

Referee comment on "Implementation of HONO into the chemistry–climate model CHASER (V4.0): roles in tropospheric chemistry" by Phuc Thi Minh Ha et al., Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2021-385-RC1>, 2022

Reviewer Comments to Ha et al.

Heterogeneous chemistry of HONO formation and sink in the atmosphere is one of the least quantified issues in tropospheric chemistry, which induce serious uncertainty in the global and regional CTM to predict O₃ formation, CH₄ lifetime, and so on. The present article implements the heterogeneous HONO chemistry into the chemistry-climate model CHASER to show the inclusion of HONO chemistry reduces the model bias against the measurements for PM_{2.5}, NO₃⁻/HNO₃, NO₂, OH, O₃ and CO in the lower troposphere.

Since the importance of heterogeneous production and loss of HONO has rarely been treated by a global CTM, it is interesting and worthwhile to evaluate the role of the chemistry of HONO in a global scale, and the present study is a challenging effort toward the direction. The most serious problem of the present article, however, is that the effects of HONO chemistry in the global atmosphere are discussed in 3.1 and 3.2 without enough validation of the processes and assumed uptake coefficients for heterogeneous formation and loss of HONO. At the present stage of understanding of heterogeneous HONO chemistry, selection of appropriate processes and uptake coefficients to reproduce the HONO concentration in the urban and remote field measurements should be the starting point for the model discussion. I am afraid that the present article does not fulfill such requirement.

Therefore, I rather reject the present version of the paper for publication, but do encourage the authors to revise the paper considering the following comments and resubmit the paper after conducting appropriate recalculation.

Specific suggestions for improving the paper:

- Many field observations and regional CTMs have revealed that the concentrations of daytime HONO in urban area is much higher than expected by models considering only dark heterogeneous formation process (e.g., Lee et al., 2016; Lu yet al., 2018; quoted in the present paper). Since photochemical heterogeneous formation is now widely accepted to be important as a source of HONO, it should be taken into account as one of the important processes in the global model as well. The present paper discusses such photochemical process only later in 3.1.5.
- Further, it has been generally accepted that every surface including soil dust and PM₅, and the earth's ground would be effective for the formation of HONO. These processes should be taken into account in the global model as well.
- As for the quantitative choice of uptake coefficients of heterogeneous processes, it is suggestive to parameterise them to reproduce the observational HONO concentration in the field. The sensitivity of uptake coefficient of the heterogeneous formation processes would be high in the boundary layer in polluted areas, validation in urban areas should be performed at first by the global model comparing with the results of regional models. Then, validation for vertical profiles by aircraft measurements of EMeRGe and Atom1 should be made before the discussion of the effect of HONO chemistry in the lower troposphere in general.
- Although the present paper refers limited laboratory studies on the heterogeneous dark reactions of HONO formation and loss on specific aerosol such as soot, the uncertainties are more than an order of magnitude, and it is not appropriate to select particular values for the "standard" run as in Table 2. Considering the large uncertainty of the uptake coefficients at this stage, the empirical approach mentioned above is suggested to be followed in the future paper.

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