Response to the Anonymous Referee #1 and #2's Reports,

We thank the Anonymous Referees for our manuscript's thorough and constructive comments.

Referee's suggestion:

The revised manuscript has been much improved by the intensified validation of the model by the observed values obtained in EMeRGe. The present version of the article is basically acceptable for publication and the following comments are for improving the paper to an advanced stage.

1. Comparison of the measurement data and the CHASER simulation for HONO/NOx in the urban/suburban boundary layer, where aerosol concentration is high and cloud contribution may be negligible. There are a few numbers of papers reporting high HONO in urban/suburban area in China and US, e.g.,

Lee et al., Atmos. Chem. Phys., 16, 2747-2764, 2016.

Ye et al., Atmos. Chem. Phys., 18, 9107-9120, 2016.

Zheng et al., Atmos. Chem. Phys., 20, 5457-5475, 2020.

Xue, et al., Atmos. Chem. Phys., 22, 1035-1057, 3149-3167, 2022.

Please show the comparison of the measured and model simulated values for HONO/NOx using the best selected common γ-values for heterogeneous HONO formation processes on aerosol- and ground-surfaces including the heterogeneous photochemical HONO formation.

Author's response:

We thank the referee for their encouragement and the suggestion of a more comprehensive order for our manuscript.

However, we may preserve the order of the verification part (Sect. 3.1), beginning with the model comparison with EMeRGe in the free troposphere to utilize the HONO-measured data on HALO. This very inclusive dataset drives our effort to make various trials on different HONO production mechanisms for understanding HONO chemistry. Only through this comparison did we notice that the standard and sensitivity simulations (Table 3) were insufficient to explain the measurement along EmeRGe. We also noticed that additional cases (Table 5), with our trials for various mechanisms, needed to explain the measurement. Several cases were successful for different conditions along EMeRGe flights, while other cases were more efficient for other comparisons (ATom, Mirai, EANET, EMEP). The order of our comparison hence started from EmeRGe (free troposphere in coastal East-Asian region) to ATom (free troposphere in the near and far-coastal regions), Mirai (coastal surface sea environment), then EANET, EMEP (continental ground-based observations). Besides EANET, EMEP, we added a comparison for HONO, NO2, O_3 during the summer of 2018 of Mt. Tai (China) by reproducing data in Xue et al.'s report (Xue et al., 2022) to compare with simulated concentrations in our model **(Lines 529 - 571)**. We then proposed the most potential HONO production mechanisms for the specific environments being compared.

Comparing CHASER simulation and the measurement in the urban/suburban boundary layer from EANET, EMEP, and Xue's data (Sect 3.1.5), we acknowledge that aerosol concentration is high for these environments. We discussed that the relative importance of aerosol uptakes in the sensitivity of HONO formation for the summit station was higher than those for the foot station **(Lines 541-544)**, and the sensitivity of O_3 formation **(Lines 559-562)**.

Referee's suggestion:

2. Next, show the comparison of measurement data of EMeRGe and model simulation demonstrating that the inclusion of heterogeneous formation of HONO on cloud water improves the agreement.

Author's response:

In the impact parts (Sect. 3.2), we added detailed discussions for aerosols and cloud effects for specific environments being compared in Sect. 3.1, concluding that adding cloud effects improved agreement in the free troposphere (generally improved for comparison with ATom, partially improved in CO simulation by cloud effect) (**lines 807-830**).

Referee's suggestion:

Discuss the **relative importance of cloud surface process for HONO formation in the free troposphere in a global scale**.

Author's response:

The relative importance of aerosol effects on HONO formation, including particle NO3-photolysis (JANO3-B and maxST+JANO3-B cases), was discussed in Sect. 3.2 (**Lines 823-830**).

The case JANO3-B and the combined case maxST+JANO3-B enhanced NO_2's aerosol-uptakes (R4, R5) and NO_3^- photolysis (R7) on the ground and aerosol surfaces (SAD threshold was set as 10^{-6} - 10^{-4} cm^2 cm^{-3} to exclude this photolysis on cloud particles). However, in these simulations, the cloud effects for the uptakes of NO_2 and HONO (R4, R6) still be active. Thus, we used this simulation to discuss the sensitivity of HONO formation and relevant chemistry to aerosol effects (not the magnitude of aerosol-effect themselves).

Regarding the relative importance of the cloud surface process for HONO formation and global oxidizing power in the free troposphere, we added in Sect. 3.2 the discussion on the magnitude of cloud effects (= GR+HR(cld) – GR; see configurations for simulations in Table 3) in **Lines 807-822**. Also, the sensitivity of HONO formation and oxidizing chemistry to cloud surface was also added in Sect. 3.2, via enhancement of (R4) on cloud surface in the ratR4+CLD case ($\gamma_{liq.}$(R4) = 0.01) and via SAD threshold for cloud in JANO3-C cases.

Referee's suggestion:

3. Decrease of HOx and O3 formation by the inclusion of HONO formation processes in the free troposphere have been discussed, which is against the general understanding that HONO formation increases HOx concentration and O3 production in urban area. If it is ascribed to the situation under low concentration region of NOx, threshold concentration of NOx, where positive to negative contribution of HONO formation to the oxidant formation will occur, should be discussed.

Author's response:

To draw a picture of the correlation between NOx concentration and HONO's impacts on oxidizing chemistry (OH, O3, CO, CH4), we added discussion in Sect. 3.2. In **lines 834-843**, the tendency sensitivity of HONO's impact on oxidant species (OH and O_3) to NO_x concentration was added. In **lines 856-872**, the discussion on the positive to a negative contribution of HONO formation mechanisms to the oxidant formation will occur was added.

Review report from Referee #2:

A second review report on "Implementation of HONO into the chemistry-climate model CHASER (V4.0): roles in tropospheric chemistry" by Phuc et al., 2021. My responses are based on the authors' responses and the revised version of the manuscript.

Since this is a revised manuscript, I don't think the manuscript is publishable since the authors still didn't account for the ground sources, which are the major HONO source, and because they calculate NO2 reduction of 20% which I don't believe a correct number since global HONO/NOx ratio can't exceed 4% even if all sources are accounted for.

Author's response:

Thank you so much for your time in reviewing our manuscript thoroughly once more. We understand that the revised manuscript did not align with your thinking.

In the revised manuscript, we tried to account for the ground source of NO_3^- photolysis (JANO3-A case), which was suggested by Lee et al. (2016). The contribution of particle NO_3^- photolysis on the ground surface (JANO3-A case) was addressed in comparison with EmeRGe (Sect. 3.1.2) and once more in the impact part (Sect. 3.2.3; **lines 847-849**).

The calculated NO_2 reduction of 20% by the STD case is naturally sceptical. After adding particle-phase NO_3^- photolysis to the model, the global impacts on tropospheric NO_x levels ranged in different tendencies (Figure 15). Only in the JANO3-C case could the strong NO3-photolysis on ground, aerosols, and cloud surfaces (SAD (10 μm2cm-3) increase the global NO_x abundance. In most of the other simulations, the NO_x abundance was still reduced due to its removal via NO_2 uptakes, followed by OH and O_3 reductions. Hence, we concluded that the impact tendencies were similar to the STD case as NO_2 uptakes are still an active removal process for NO_x, but the magnitude of global changes in tropospheric species are still with high uncertainties.

Reference

Xue, C., Ye, C., Kleffmann, J., Zhang, C., Catoire, V., Bao, F., Mellouki, A., Xue, L., Chen, J., Lu, K., Zhao, Y., Liu, H., Guo, Z., and Mu, Y.: Atmospheric measurements at Mt. Tai – Part I: HONO formation and its role in the oxidizing capacity of the upper boundary layer, Atmos. Chem. Phys., 22, 3149–3167, https://doi.org/10.5194/acp-22-3149-2022, 2022a.

Sincerely,

On behalf of all co-authors,

Phuc T. M. Ha.