

General comments:

Tropospheric chemistry often begins with a trace gas molecule reacting with one of the tropospheric oxidants: OH radicals, NO₃ radicals, or O₃ molecules. (Reactions initiated by Cl atoms, generated from ClNO₂ photolysis, also contribute to the atmosphere's oxidising capacity, although Cl chemistry was not considered in this paper.) Such reactions determine the lifetime of trace gases in the atmosphere; this chemistry also leads to the production of secondary pollutants such as tropospheric ozone, organic nitrates (PAN etc) and secondary aerosol particles. Under most conditions, OH radicals make the largest contribution to the atmosphere's oxidising capacity. However there are gaps in our detailed understanding of OH sources, notably concerning the importance of the photolysis of nitrous acid (HONO).

This paper reports observations of HONO made in an urban location (Tai'an city) in the central North China plain over a period of several weeks in summer. The authors find that simplest "default" HONO source from the OH + NO reaction (which is not a net source of OH) only accounts for 13% of the observed HONO, averaged over the measurement campaign. Thus a large additional "unknown" HONO source must exist, and the authors explore 6 possibilities. The dominant HONO source at the measurement site is identified as the heterogeneous conversion of NO₂ into HONO on ground surfaces, either photo-enhanced by sunlight during the day (approx. 80% of the source strength), or without light in the dark at night (approx. 70%). Small contributions (of up to around 10% each) variously come from the conversion of NO₂ into HONO on aerosol during the day or at night, the photolysis of particulate nitrate, or direct emissions of HONO (e.g. from traffic or soils). The authors have done a good job to pick apart and characterize the strengths of these different possible HONO sources. This information is valuable to atmospheric chemists and modellers.

In the second half of the paper, the authors employ a zero dimensional box model to investigate what contribution OH derived from HONO makes to the radical chemistry at their measurement site. The model is used to model OH from HONO and from other sources (ozone photolysis, unsaturated VOCs reacting with ozone, etc), sources & sinks of HO₂ and RO₂ radicals, sources & sinks of NO₃, and the changing contributions of OH- and NO₃-initiated oxidation chemistry over the 24 hour cycle. The model's many outputs are nicely illustrated on well-constructed and informative plots. But this part of the work was less convincing due, primarily, to the simplicity of the 0-D model which cannot consider the vertical gradients in HONO concentrations that are known to exist from ground-based HONO sources. The authors are honest about the limitation of their modelling approach and say that a 1-D box model would provide better conclusions. The nighttime oxidant NO₃ and its reservoir N₂O₅ were not measured in this study, but modeled instead, and this leads to further uncertainties in assessing the relative contributions of OH and NO₃ reactivity (as acknowledged by the authors).

[Response: Thanks for your great efforts and valuable comments, which helps to improve our manuscript. Please see the point-to-point response below.](#)

(Comments in Black, Response in Blue, Changes in the manuscript in Red)

Specific comments:

[Comment 1] The success (or otherwise) of the 0-D box model depends on making the right choice for HONO's mixing layer height (section 3.2.2.4, line 343). The authors choose 50 m for HONO's MLH because that generates the best agreement between modeled and measured HONO production rates, P(HONO), in Fig 6(b). But Figure 6(b) shows that setting MLH to 35 m instead of 50 m

increases P(HONO) by roughly a factor of 1.5; alternatively, setting MLH to 100m decreases P(HONO) by a factor of 2. Thus relatively small changes in the MLH generate big changes in modeling the ground HONO source (i.e. the dominant HONO source), with implications for understanding the OH source strength and radical chemistry. This raises questions that require answers:

* How much do this paper's conclusions depend on the precise choice of the MLH? – how would the conclusions change if the actual MLH were 35 m or 100 m or 200 m?

Response: We have to admit that MLH has a strong impact on modeling ground-derived HONO sources in a box model, for which a homogeneous MLH is assumed, i.e., with the same kinetics, an increasing MLH would cause a decreasing contribution of a ground-derived source. However, MLH does not affect a) aerosol-derived HONO sources (see the equations for related parametrizations in Sections 3.2.2.2 and 3.2.2.3), assuming no significant gradient of the particle concentration in the MLH or b) the main conclusions of this paper. Please see the detailed explanation in the response to the followed comment below.

* Is there a way to independently verify that the optimum choice of HONO's MLH is 50 m? – can this MLH value be proved by comparing HONO observations made at the foot site in Tai'an city with HONO measurements at the nearby summit of Mount Tai (using data from the companion paper)? Is the vertical gradient in HONO at this site consistent with other studies e.g. Brown et al (J GeoPhys Res, 118, 8067, 2013; doi:10.1002/jgrd.50537) who also observed vertical gradients in NO₃, N₂O₅ and ClONO₂?

Response: Due to the gradients of HONO observed in the atmosphere, near-ground surface HONO measurements are typically more weighted by ground-derived sources (Vandenboer et al., 2013; Vogel et al., 2003; Wong et al., 2011, 2013). As shown in a recent vertical HONO measurement in southwest China (Xing et al., 2021), HONO levels rapidly decreased from 4.8 ppbv at the ground level (~4 m above the ground surface) to 1.6, 0.7, 0.3, 0.2, and 0.1 ppbv averaged in height ranges of 0 – 100, 100 – 200, 200 – 300, 300 – 400, and 400 – 500 m above the ground level, respectively. This indicates that the near-ground HONO was remarkably affected by ground-derived sources and hence the MLH should be much less than 100 m in box model studies constrained by near-ground surface measurement data. A similar phenomenon could also be found in Brown et al. (2013) and Vandenboer et al. (2013) in which they used a tower-based platform to measure the vertical profile of HONO. Therefore, we chose 50 m as the MLH.

Comparison between HONO measurements at the foot and the summit could not help constrain MLH because HONO at the summit station was mainly originated from the vertical transport driven by mountain winds rather than typical atmospheric vertical turbulences (see the companion paper about the summit observations at <https://acp.copernicus.org/preprints/acp-2021-529/#discussion>).

To reduce the uncertainties caused by the assumed MLH, we also conducted a) several sensitivity tests with different MLH (35, 50, and 100 m, see Figure 6B and Section 3.2.2.4) and b) enlarged aerosol-derived sources to see whether the aerosol-derived sources could explain the observations if ground-derived sources were reduced. Results showed a significant discrepancy between the modeled HONO and the observations assuming aerosol-derived sources dominated HONO formation (Figures S8 and S9), which reinforced our main conclusion about the HONO budget.

Hence, the MLH used in this study may have uncertainties (honestly, this is still an open question for near-ground HONO studies), but does not change the main conclusions about the HONO budget.

Additionally, we also addressed the necessity of the vertical measurement based on tower-, balloon-, or aircraft-based platforms and the coupling with 1-D model simulations (See Section 3.2.2.4).

* How does HONO's MLH change with e.g. wind speed, time of day, daytime versus night? Was a constant MLH = 50 m assumed to model the HONO time series (Fig 7a) and HONO's average diurnal profile (Fig 7b)? How would variability in MLH affect this work's conclusions?

Response: HONO's MLH may vary with wind speed, time of day, daytime versus night, etc. however, without continuous vertical measurement, the variation of HONO's MLH could not be achieved. Please see the response to the previous comment for the use of MLH and related work we did to reduce its uncertainties.

A constant MLH (50 m for normal simulations and 35 or 100 m for sensitivity tests) was used for all the model simulations, including Figures 7a and 7b.

Please see the above response for the impact of MLH variability in the conclusions.

[Comment 2] Figures 1 and 2 show detailed time series of the observations, and Table 2 provides statistics (all good). The information that is missing is average diurnal profiles of NO, NO₂, O₃, J(HONO) etc – please add these plots either in section 3.1.1 or the supporting information. Please add the diurnal profile of J(HONO) to one or all of Fig 4, Fig 6 and Fig 7(b) [and Fig S7(b) and Fig S8(b)], so that reader can easily distinguish day vs night, sunrise and sunset, photolytic vs dark HONO sources.

Another reason why I request the diurnal plots of NO, NO₂, O₃ and J(HONO) etc is because the diurnal plot of P(HONO) is not symmetric around 12:00 noon (Fig 4 and Fig 6). The unknown HONO source is faster in the morning than the afternoon, and peaks at 11am. The authors note this asymmetry on line 259 but they don't explain it. What is the cause of this asymmetry in HONO? And is P(HONO) expected to also be asymmetric at other locations? [I'm guessing it's not due asymmetry in J(HONO) because P(OH) from ozone photolysis *is* symmetric around 12:00 noon in Fig 14c.]

Response: We added diurnal variations of HONO, O₃, NO₂, J(NO₂), PM_{2.5}, CO, NO, and SO₂ in the supporting information (Figure S3) and J(HONO) in Figure 4, which are shown below:

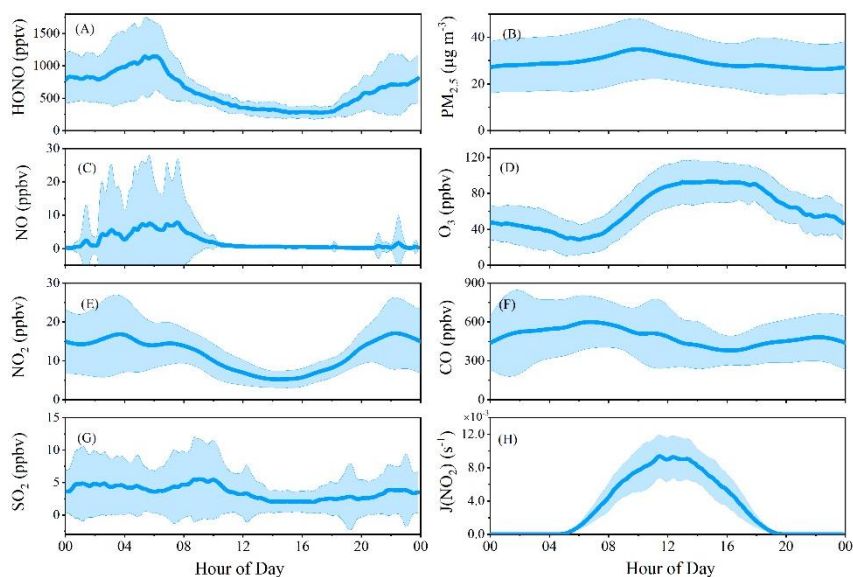


Figure S3: Diurnal profiles of HONO and related species measured during this campaign. Note that

some of these data were also shown in the companion ACP paper for comparison between measurements at the foot and the summit stations.

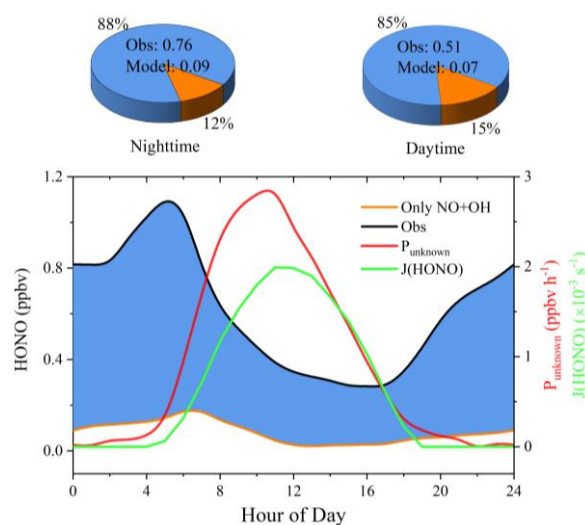


Figure 4: Simulated HONO by the default mechanism (Sce-1, left axis) compared with the observations (Obs, left axis), unknown source strength (P_{unknown} , right axis), HONO photolysis frequency ($J(\text{HONO})$, right axis), and the relative contributions of NO + OH to the observations at night (19:00 – 4:50, left pie chart) and day (5:00 – 18:50, right pie chart), respectively. The shaded area in blue represents the difference between the observation and modeled values.

Regarding the asymmetric variation of HONO and P_{unknown} , in Section 3.2.1, we improved the related text as “Note that the profile of P_{unknown} was asymmetric around 11:00, indicating the unknown source is not simply photolytic but also includes its precursors that also have an asymmetric distribution (e.g., NO_2 , Figure S3).” Besides, we found that the scenarios with enlarged aerosol-derived sources showed worse performance in reproducing this asymmetry, which could be another piece of evidence that aerosol-derived sources are not the dominant ones. We then improved related text in Section 3.2.3 as “While that the modified model could also generally predict the observed HONO levels (Figures S8A and S9A) but it largely failed to reproduce the noontime observations in levels and variations (Figures S8B and S9B) including its asymmetry as mentioned in Section 3.2.1. This observation reinforces our conclusion that aerosol-derived sources play only a minor role in the daytime HONO formation.”

[Comment 3] How do the conclusions of this paper translate to other locations in China and other countries? In terms of air quality, this is a polluted site, so are conditions here somehow special? For example, because high $[\text{NO}_2]$ produces an especially large ground-based HONO source at this site? And/or because high ozone, $[\text{O}_3] > 100 \text{ ppbv}$, is reacting rapidly with unsaturated VOCs to produce an especially active HO_2 and RO_2 chemistry at this site? For example, NO_3 is traditionally seen as the dominant nighttime oxidant, yet the modeling here in this study shows the sink for isoprene (C_5H_8) at night is approximately 50% to OH and 50% to NO_3 – it is unusual to see such active OH reactions at night. [BTW: I agree with the authors that it is better to measure NO_3 than to model it.]

Response: Generally, we believe that the parametrizations for HONO sources could be broadly used for near-ground measurements worldwide and similar conclusions are also expected in polluted

regions. For instance, our previous study (Xue et al., 2020) found similar conclusions in a polluted site in the winter North China Plain, where air pollution (aerosol, NO₂, HONO, etc.) is much higher than that at this site. It is noteworthy that the method for quantifying direct HONO emission, which is firstly proposed in this study, could significantly reduce the overestimation of direct HONO emission during daytime. We suggest further studies to use this method to have more accurate results. At this site, high O₃ levels were observed both during the daytime and the night-time (Figure S3D), which is typical for the increasing O₃ pollution in the North China Plain (Han et al., 2020; Li et al., 2019; Sun et al., 2016, 2019). Hence reactions of O₃ with unsaturated VOCs play important role in OH production, particularly during the night-time maintaining and dominating OH production (Figures 9A and 13A).

As OH and NO₃ measurements are not available, we can only use the model results to shed light on the atmospheric chemistry in this polluted region and point out the uncertainties. A related text in Section 3.3.3.2 was improved as “Model results may have uncertainties but shed light on the atmospheric chemistry in this polluted region. By far, very few NO₃ measurements are available in China (Lu et al., 2019; Suhail et al., 2019), while its high concentration and important role in chemical oxidation presented in this study indicate the necessity of direct NO₃ (as well as related species such as N₂O₅, ClNO₂, etc.) measurements in the NCP, where summertime O₃ levels are substantially increasing (Han et al., 2020; Li et al., 2019; Sun et al., 2016, 2019).”

Technical corrections:

Generally, the paper is written well. However, some of the sentence structure is long and complex, especially those sentences that list multiple measurements, ideas or findings. Consider rewording and/or breaking into separate sentences [e.g. lines 40-43; line 132 (see below)].

Response: We separated Lines 40-43 as two sentences: “HONO photolysis in the early morning initialized the daytime photochemistry at the foot station. It was also a substantial radical source throughout the daytime, with contributions higher than O₃ photolysis to OH initiation.”

Line 132 was improved as “10-min average data (except for PM_{2.5}) were used for the analysis of time series and statistic descriptions of the data. In contrast, hourly data were used for model simulations.”

In addition, the English of the manuscript was carefully improved by the authors with the help of comments from three reviewers (See the response to comments and the manuscript).

Some figure captions also have long and complex sentence structure; they likewise could be improved by breaking them into several shorter sentences, e.g. Fig 9 “Simulated concentrations of (A) OH... and (D) NO₃. [New sentence] The different coloured lines show...”.

Response: Captions for Figure 9 were modified as “Simulated concentrations of (A): OH, (B): HO₂, (C): RO₂, and (D): NO₃. Different colored lines show results in different scenarios (NO + OH: only with the homogeneous source; Base: constrained by the observed HONO; +50%: constrained by the observed HONO×1.5; +100%: constrained by the observed HONO×2; -50%: constrained by the observed HONO×0.5; -100%: constrained by HONO = 0).”

Besides, other figure captions were also improved similarly as the reviewer suggested.

Fig 13: the 3 notes in the figure caption would work better as 3 separate sentences.

Response: Improved as suggested.

Fig 4: caption needs separate sentences to explain the main panel and the pie charts.

Response: Improved as suggested.

Abstract line 32: “Our model’s default HONO source from the OH + NO reaction could only reproduce 13% of the observed HONO...”

Response: Improved as suggested.

Abstract line 37: “A HONO/NO_x ratio of 0.7% from direct emissions [plural] was inferred...”. Also please state what are the direct sources of HONO at this measurement site?

Response: This line was improved as “A $\Delta\text{HONO}/\Delta\text{NO}_x$ ratio of 0.7% for direct emissions from vehicle exhaust was inferred and a new method to quantify its contribution to the observations was proposed and discussed.”

Abstract line 43: State clearly that NO₃ concentrations come from the 0-D model. Otherwise, without reading the main body of the paper, readers might assume NO₃ was measured.

Response: We added “modeled” before NO₃.

Abstract lines 46-48: the sentence repeats itself.

Response: the repeated part was deleted. “At night, NO₃ chemistry led to 51% or 44% of P(HNO₃) or the C₅H₈ oxidation, respectively. ~~NO₃ chemistry may significantly affect night-time secondary organic and inorganic aerosol formation in this high-O₃ region,~~ implying that NO₃ chemistry could significantly affect night-time secondary organic and inorganic aerosol formation in this high-O₃ region.”

Line 79: NO₃ radicals [plural]

Response: Improved as “the NO₃ radical”.

Line 99: However, very few observations have been reported of OH and NO₃ concentrations...

Response: Improved as suggested.

Line 115: Mt Tai is located in...

Response: Improved as suggested.

Line 117 / end of section 2.1.1: Add a sentence to explain the scientific motivation for making HONO observations at this ground site and at the summit of Mt Tai, and briefly explain what the authors found. Readers should be able to know this without having to find and read the companion paper.

Response: Thanks for the suggestion. We added a sentence: “Measurements at these two stations allow us to study HONO formation and its role in the atmospheric oxidizing capacity of the lower (the foot study) and the upper boundary layer (the summit study). Briefly, in the summit study, we found remarkably high daytime HONO levels as well as high unknown HONO source strength, which was mainly caused by rapid vertical transport from the ground to the summit levels driven by mountain winds.”

Line 120-121: Here it states the LOPAP measurements took place in 2017. But line 109 and the time axis labels in Fig 1 and 2 show 2018. Which is correct?

Response: Sorry about this mistake. The campaign was taken place in the summer of 2018. We corrected the text in the manuscript.

Line 132: Reword: 10 minute data were used in X. [New sentence] Hourly data were used in Y.

Response: Improved as suggested.

Line 133: PM2.5 measurements were obtained... [plural]

Response: Improved as suggested.

Line 136: SZA functions [plural]

Response: Improved as suggested.

Line 144: interferences in [not “of”] the NO₂ measurements [plural]

Response: Improved as suggested.

Line 169-171: Air masses [plural] observed at this site [delete “was”] originated from... which are shown in the wind rose plot in Figure S2.

Response: Improved as suggested.

Line 256: HONO mixing ratio differences... were calculated... and they are compared with P_unknown in Figure S5. [plurals]

Response: Improved as suggested.

Line 257: What do the authors mean by the word “typical”? – the average diurnal profile of P_unknown inferred from their HONO observations? I doubt P_unknown shown in Fig 4 can typically be applied to all other measurement sites.

Response: The sentence was deleted.

Line 278: Add a reference to the recent study of HONO emissions from traffic in a road tunnel by Kramer et al, who found a similar HONO/NO_x ratio of 0.85%. <https://doi.org/10.5194/acp-20-5231-2020>.

Response: Thanks for the information. We then added this reference.

Line 291 / Table 4: Why are there two values listed for HONO (and two values for NO and two values for NO_x)? Are they the HONO concentrations at the start and end of each plume of fresh emissions? Also units = ppbv?

Response: We added a sentence in the caption of Table 4: “Concentrations (ppbv) of HONO/NO/NO_x at the start and the end of each plume of fresh emissions are also shown.”

Line 302: What is meant by “overestimated one” and “popularly used one”?

Response: The “overestimated one” refers to the γ_a value inferred from the summit study. Briefly,

we assumed that all the unknown HONO sources originated from NO₂ uptake on the aerosol surface then we could get a γ_a value that constrained its upper limit (details can be found in the companion paper). The “popularly used one” refers to the γ_a value obtained from laboratory flow tube experiments (Stemmler et al., 2006, 2007), which is popularly used in model studies.

We added related references in the manuscript.

Line 340: “reflected by” is not the right verb. [I’m not sure what the correct verb should be because I didn’t understand this sentence.]

Response: “reflected by” was replaced by “described by”.

Line 355: Therefore, the MLH for HONO was set at 50 m, with sensitivity tests performed with the MLH set at 35 and 100 m.

Response: This sentence was improved as “Therefore, the MLH for HONO was set at a constant height of 50 m, with sensitivity tests performed with the MLH set at 35 and 100 m.”

Line 371: the model with the present HONO source parameterizations performed well in predicting... [the original adjective “magnificent” is too strong].

Response: Improved as suggested.

Line 378: reinforcing our conclusion that aerosol-derived sources played only a minor role in daytime HONO formation.

Response: Improved as suggested.

Line 400: rather than “The former one...”, it is clearer simply to begin the sentence with “HONO emissions from soils may occur...”

Response: Improved as suggested.

Line 447 / Fig 10: I found it difficult (& sometime impossible) to distinguish the different OH sources and sinks, where two shades of the same colour are plotted next to each other. The rainbow of colours is visually pretty, but the plot is impossible to interpret. Likewise P(NO₃) and L(NO₃).

Response: Figure 10 and its caption were improved as follows:

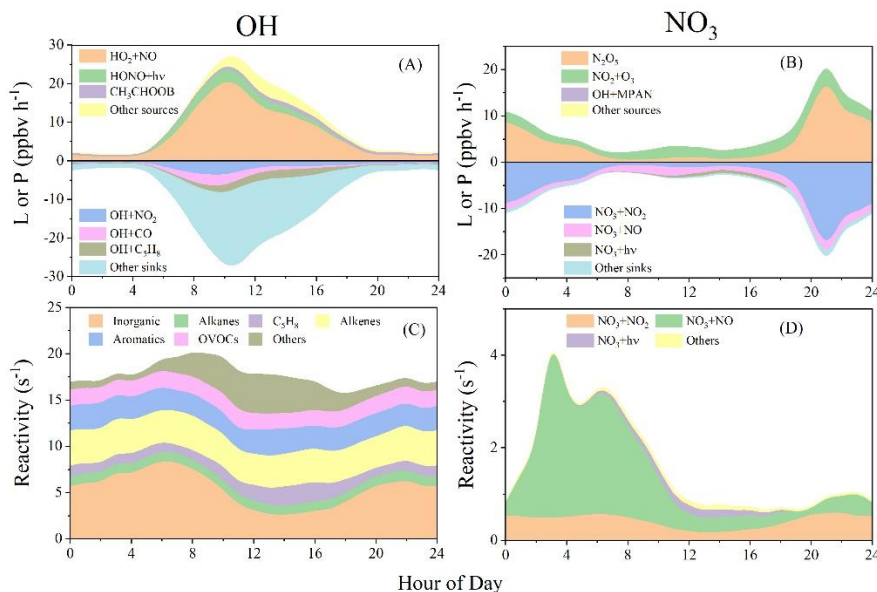


Figure 1: Production rates (P), loss rates (L) and reactivities of radicals. (A): L and P of OH; (B): L and P of NO₃; (C): Reactivities of OH and (D): Reactivities of NO₃. In (A) and (B), the top-3 sources or sinks are shown, and all the others are summarized in “Other sources” or “Other sinks”. In (C), OH reactivities with different families of the measured species are shown and reactivities with all the unmeasured species are summarized in “Others”. In (D), NO₃ reactivities from top-3 reactions are shown and all the others are summarized in “Others”.

Line 447 / Fig 10: Why is NO₃ photolysis (during the day) not plotted in the NO₃ reactivity plot, panel (D)? Or is photolysis included in the “inorganic” reactions (orange colour)?

Response: Figure 10 was improved (see above) and NO₃ photolysis was shown in the graph.

Line 476: hemiterpene emitted by very many species of vegetation...

Response: Improved as suggested.

Line 483 / section 3.3.3.1: In addition to forming particulate nitrate, it should also be noted that HNO₃ formation from OH+NO₂ and from N₂O₅ hydrolysis on aqueous aerosol are also the major daytime and nighttime sinks for removing NO_x from the atmosphere. Without these NO_x sinks, NO_x photochemistry would produce tropospheric ozone even more rapidly.

Response: We added this sentence: “HNO₃ formation from OH+NO₂ and from N₂O₅ hydrolysis on the aqueous aerosol surface are also the major daytime and night-time sinks for removing NO_x from the atmosphere.”

Line 487: the dominant one

Response: Improved as suggested.

Line 556: The default HONO source, NO + OH, significantly underestimated the observed HONO concentrations. [New sentence] This reaction could only account for 13% of the observed HONO, revealing a strong...”

Response: Improved as suggested.

References

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