We appreciate the efforts that the two anonymous reviewers and the editor have made toward improving our manuscript. Please see the point-to-point response below.

1. Response to RC1

The manuscript presents results of a comprehensive field campaign at two different altitudes, the foot (150 m a.s.l.) and the summit (1534 m a.s.l.) of Mt. Tai (Shandong province, China). Mt. Tai locates in the middle of the NCP with a relatively high pollution level. The measured HONO diurnal profile shows a daytime peak at 12:30 local time, which is interesting since HONO diurnal profiles would typically peak during the night and early morning in more polluted regions. The topic is of interest to the scientific community and is suitable for publication in ACP after addressing the comments below.

The authors claim that OH+NO gas-phase reaction accounts for only 8% of measured HONO, and that 70-98% of the unknown HONO sources can be attributed to vertical transport from ground surfaces. However, the authors didn't show/present the OH values used to calculate the OH+NO reaction rate, and they didn't consider this reaction when calculating the net production of OH from HONO. The authors used an unjustified circular assumption that OH loss in the OH+NO reaction at the ground will be recycled back to OH at a higher altitude without any valid calculation of HONO lifetime vs transport time from the ground to the summit. The authors claim that they calculated HOx budget, although they only calculated gross HONO photolysis and O3 photolysis. (primary sources of OH only). I suggest the authors limit their discussions to HONO sources and sinks, and that they should account for NO+OH reaction in calculating HONOpss or assume several OH values around those published earlier to calculate their uncertainties. Otherwise, the manuscript is publishable after addressing these comments.

Response: Thanks for your efforts and comments, which help to improve our manuscript. Please see the point-to-point response below (**Comments in Black; Response in Blue; Changes in Red**).

Specific comments:

Page 15, Line 320: The authors didn't justify the use of OH-j(O^1D) correlation from previous publications to calculate OH in this study. Although some studies showed a good correlation, it still may not be a good proxy for OH given the large variation in the obtained slops. The authors use a circular argument that OH is not important since NO+OH is not important, to justify the uncertainty associated with their approach. At which OH levels does the NO+OH reaction accounts for 8%?

Maybe, it is safer to either simulate OH using a box model or use a range of OH levels around those reported previously by Kanaya et al. (2009) to show that it is not important, as they claim This is a major issue that the authors need to address before continuing with their calculations of unknown HONO sources.

The authors used several assumptions to calculate the contribution of different HONO sources to measured HONO levels. Most importantly is the photolysis of pNO3, for which the authors used a range of enhancement factors (EF) that ranges from 1 to ~15.6, accounting for 0.6 to 9.6%, depending on EF, leaving ~93% of HONO unknown sources unknown. I think. A major uncertainty here is related to HONOpss, which the authors didn't sufficiently address, which affects the unknown fraction HONO.

Response: We agree that the estimated OH could result in some uncertainties in calculation on unknown HONO sources and net OH production. We added OH sensitivity tests and found very small impacts on $P_{unknown}$. Figure S6 and the below texts are added in the manuscript.

The estimated OH could lead to some uncertainties. Hence, we added OH sensitivity tests to reinforce our analysis and conclusion. The used OH, the corresponding $HONO_{pss}$, P_{un} and results from the sensitivity tests were also shown in Figure S6. The estimated OH level was lower than that measured during the MTX campaign (Kanaya et al., 2013). This is mainly caused by lower $J(O^1D)$ resulting from frequent cloudy weather during the present study period. For instance, the average RH during this campaign was 96%, which is much higher than that during the MTX campaign (67%). The variation of OH levels indeed remarkably impact $HONO_{pss}$. However, $HONO_{pss}$ (5-15 pptv level) is still 1-2 orders of magnitude lower than the observed HONO (50-200 pptv level), leading to a negligible impact of variable OH and $HONO_{pss}$ levels on P_{un}.

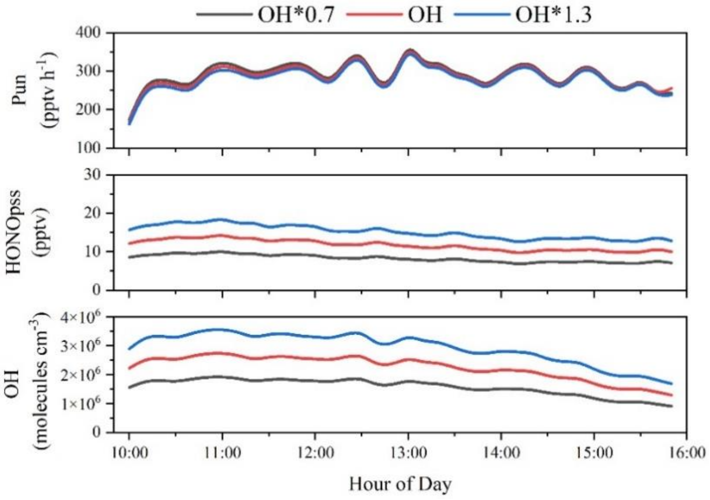

Figure S6: Estimated OH concentrations (red line) used in this study and corresponding $HONO_{pss}$ and P_{un} (red lines). Black lines represent OH level reduced by 30% and corresponding $HONO_{pss}$ and P_{un}. Blue lines represent OH level enlarged by 30% and corresponding $HONO_{pss}$ and P_{un}.

Page 18, lines 404-412: The authors' argument of OH recycling via HONO photolysis as the source of OH at higher altitude is not justified and is flawed. The authors didn't provide information about the HONO lifetime vs the transport time to this altitude. I think this whole paragraph should be just deleted.

Response: The maximum of average diurnal J(HONO) is 8.0×10^{-4} s^{-1} (Figure 9), corresponding to a minimum HONO lifetime of about 21 min against photolysis, longer than the estimated transport time of 7-17.5 min. As shown in Figure 9, the remaining proportion of HONO after a period of transport from the ground to the summit levels is about 50-80% at noontime. α could be even larger because the calculation only considers HONO loss, whereas HONO production during the transport along the mountain slope was not taken into consideration. Then whether the transport of HONO could constitute an OH transport path depends on the amount of OH consumption to produce HONO through NO+OH at the foot station.

At the foot station, NO+OH contributed 15% of daytime HONO formation and photo-enhanced NO_2 uptake on the ground surface dominated the rest as reported in the companion ACP paper (Xue et al., 2021). Besides, hydrogen peroxide (H_2O_2), an important OH reservoir, could also be transported from the ground to the summit levels as reported in our recent study (Ye et al., 2021). At the ground level, H_2O_2 was mainly produced by HO_2+HO_2 (Ye et al., 2021). Hence, it could be preliminarily inferred that radicals (i.e., OH and HO_2) could be transported through their precursors/reservoirs (like HONO and H_2O_2) with lifetimes longer than themselves.

We improved the texts as:

Radicals, including OH and HO_2, are not expected to be transported far due to their short enough lifetimes (<1 s). However, 15% of daytime HONO was formed at the ground level through NO + OH as reported in the companion ACP paper (Xue et al., 2021), and part of OH consumed at the ground level would be released at the summit level through HONO photolysis. This could be supported by our recent finding that hydrogen peroxide (H_2O_2), an important OH reservoir, could be transported from the ground to the summit levels (Ye et al., 2021). At the ground level, H_2O_2 was mainly produced by HO_2+HO_2 (Ye et al., 2021). Hence, it could be preliminarily inferred that radicals (i.e., OH and HO_2) could be transported through their precursors/reservoirs (like HONO and H_2O_2) with lifetimes longer than themselves. Furthermore, the enhanced vertical air mass exchange could also lead to fast transport of other pollutants ($PM_{2.5}$, O_3, CO, SO_2, etc.) from the ground to the summit levels, which will significantly impact the atmospheric composition as well as its chemistry in the upper boundary layer or the residual layer. The discussion and implications in this study are instructive for further laboratory or model studies.

Page 18, line 414: provide a reference…

Response: A reference was added.

(Jiang et al., 2020)

Page 19, lines 418-420: This long sentence is not clear at all….either provide all relevant information or leave it for the accompanying paper. Otherwise, HONO net photolysis should be used to calculate HONO relative contribution to OH primary sources.

Page 19, lines 428-438: Again, this is all irrelevant if HONO net contribution is not calculated.

Response to both comments: The whole of Section 3.6 was improved as we replaced gross OH production from HONO photolysis with its net OH production (Figure 11). We also calculated the contribution $P(OH)_{HONO\_net}$ to $P(OH)_{sum}$ (Figure S8).

The improved figures and Section 3.6 are as follows:

3.6 Role of HONO in the Oxidizing Capacity of the Lower and the Upper Boundary Layer

O_3 was typically the major OH source at high altitude regions, including the upper boundary layer. Then we compared the OH production rates from O_3 and HONO photolysis to investigate whether HONO could play a significant role in the oxidizing capacity of the atmosphere at this high-altitude site. Photolysis of HONO and O_3 with their net OH production is shown in R-2 and R-5 to R-7, respectively. OH loss through HONO + OH and NO + OH was subtracted from $P(HO_x)_{HONO}$ to obtain $P(HO_x)_{HONO\_net}$.

$\boldsymbol{O_3 + h\nu \rightarrow O_2 + O(^1D),}$ **J(O(¹D))** **R-5**

$\boldsymbol{O(^1D) + H_2O \rightarrow 2OH,}$ **k_3** **R-6**

$\boldsymbol{O(^1D) + M\ (N_2\ or\ O_2) \rightarrow O(^3P) + M\ (N_2\ or\ O_2),}$ **k_4** **R-7**

$$\boldsymbol{P(HO_x)_{HONO\_net} = [HONO] * J(HONO) - k_1 * [NO] - k_2 * [HONO],} \qquad \textbf{Eq-8}$$

$$\boldsymbol{P(HO_x)_{O_3} = [O_3] * J(O(^1D)) * \phi,} \qquad \textbf{Eq-9}$$

where the reaction constants were taken from the IUPAC kinetic database (https://iupac-aeris.ipsl.fr). The atmospheric RH and temperature largely influenced the branching ratio of R-6 toR-7. The average OH yield (ϕ) during the campaign of 20% was used for calculating OH production from O_3 photolysis.

Additionally, in the companion paper in which HONO was reported to be the most important primary OH source at the foot station (Xue et al., 2021). A comparison between the role of HONO at the foot and the summit stations could provide more insights into the importance of HONO throughout the boundary layer. Moreover, as reported in the companion paper, HONO observed at the foot station was mainly produced through NO_2 heterogeneous reactions and NO+OH. Therefore, the comparison could also shed light on the link between the atmospheric oxidizing capacity in the lower and the upper boundary layer, although measurements at two stations were conducted during two consecutive periods rather than the same one in summer 2018.

Figure 11 displays the diurnal profiles of net OH production rates from HONO and O_3 photolysis at the foot and the summit stations. It is apparent that both $P(OH)_{HONO\_net}$ and $P(OH)_{O_3}$ showed higher levels at the foot station compared to the summit station. For instance, average $P(OH)_{HONO\_net}$ and $P(OH)_{O_3}$ at the foot station are 0.9 and 0.5 ppbv h^{-1}, respectively, both of which are significantly higher than those (0.06 and 0.28 ppbv h^{-1}) at the summit station. This is caused by relatively lower HONO and O_3 concentrations and lower solar photolysis frequencies as a result of frequent cloud formation observed at the summit station.

In particular, after night-time accumulation, HONO photolysis is found to initialize daytime photochemistry in the early morning at the ground level (Alicke et al., 2002; Kleffmann, 2007; Platt et al., 1980). This was also observed at the foot station. As shown in Figure S8, at the foot station, the contribution of $P(OH)_{HONO\_net}$ to $P(OH)_{sum}$ was almost 100% at sunrise around 5:00. It showed a declining trend but still played the dominant role in $P(OH)_{sum}$, with a contribution larger than 90% in the early morning (5:00-7:00). At the summit station, at 5:00, solar radiation was very weak, for instance, $J(NO_2)$ was only 3.6×10^{-4} s^{-1}. At this time, $P(OH)_{HONO\_net}$ was slightly negative (-7×10^{-3} ppbv h^{-1}) due to OH loss through HONO + OH and NO + OH. O_3 photolysis was initialized at the same time, but $P(OH)_{O_3}$ was nearly zero (7×10^{-4} ppbv h^{-1}). From 6:00 to 7:00, a considerable amount of net OH was produced through HONO photolysis (0.04-0.09 ppbv h^{-1}), with its contribution to $P(OH)_{sum}$ decreasing from 64% to 39% (Figure S8). Hence, it could be inferred that daytime atmospheric photochemistry at the summit level is also initialized by HONO photolysis.

On average, the contribution of $P(OH)_{HONO\_net}$ to $P(OH)_{sum}$ was 64% at the foot station, higher than that (18%) at the summit station (Figure 11), indicating the essential role of HONO in the atmospheric oxidizing capacity at both the ground (lower boundary layer) and the summit (upper boundary layer) levels in mountainous regions. As discussed before, the transport from the ground to the summit levels contributed to the majority of HONO observed at the summit level. This points to a new insight that ground-derived HONO played an important role in the oxidizing capacity, not only at the ground level but also in the upper boundary layer (~1500 m) in mountainous regions. Yet this vertical exchange might be only valid in the mountainous areas, and the follow-up regional impact still needs to be quantified by further model studies.

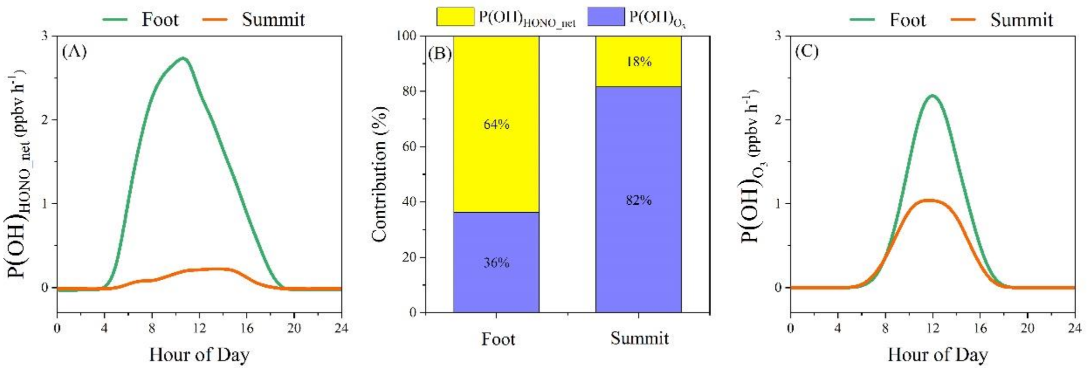

Figure 11: OH production from photolysis of HONO ($P(OH)_{HONO\_net}$) and O3 ($P(OH)_{O_3}$) at the foot and the summit of Mt. Tai. (A): $P(OH)_{HONO\_net}$, (B): relative contributions, and (C): $P(OH)_{O_3}$.

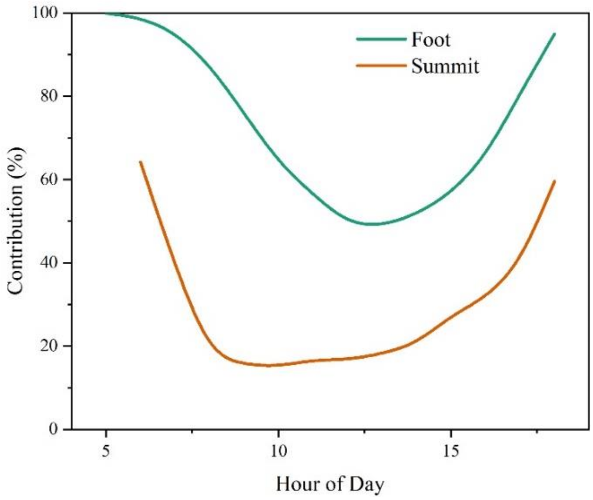

Figure S8: Relative contribution of $P(OH)_{HONO\_net}$ to $P(OH)_{sum}$ at the foot and the summit stations.

Page 19, lines 425-445: replace HOx with OH since you HONO and O3 photolysis are sources of OH only, not HO2.

Response: Done.

Reference

Alicke, B., Platt, U. and Stutz, J.: Impact of nitrous acid photolysis on the total hydroxyl radical budget during the Limitation of Oxidant Production/Pianura Padana Produzione di Ozono study in Milan, J. Geophys. Res. Atmos., 107(D22), 8196, doi:10.1029/2000JD000075, 2002.

Jiang, Y., Xue, L., Gu, R., Jia, M., Zhang, Y., Wen, L., Zheng, P., Chen, T., Li, H., Shan, Y., Zhao, Y., Guo, Z., Bi, Y., Liu, H., Ding, A., Zhang, Q. and Wang, W.: Sources of nitrous acid (HONO) in the upper boundary layer and lower free troposphere of the North China Plain: insights from the Mount Tai Observatory, Atmos. Chem. Phys., 20(20), 12115–12131, doi:10.5194/acp-20-12115-2020, 2020.

Kanaya, Y., Akimoto, H., Wang, Z. F., Pochanart, P., Kawamura, K., Liu, Y., Li, J., Komazaki, Y., Irie, H., Pan, X. L., Taketani, F., Yamaji, K., Tanimoto, H., Inomata, S., Kato, S., Suthawaree, J., Okuzawa, K., Wang, G., Aggarwal, S. G., Fu, P. Q., Wang, T., Gao, J., Wang, Y. and Zhuang, G.: Overview of the Mount Tai Experiment (MTX2006) in central East China in June 2006: Studies of significant regional air pollution, Atmos. Chem. Phys., 13(16), 8265–8283, doi:10.5194/acp-13-8265-2013, 2013.

Kleffmann, J.: Daytime Sources of Nitrous Acid (HONO) in the Atmospheric Boundary Layer, ChemPhysChem, 8(8), 1137–1144, doi:10.1002/cphc.200700016, 2007.

Platt, U., Perner, D., Harris, G. W., Winer, A. M. and Pitts Jr, J. N.: Observations of nitrous acid in an urban atmosphere by differential optical absorption, Nature, 285(5763), 312–314, doi:10.1038/285312a0, 1980.

Xue, C., Ye, C., Kleffmann, J., Zhang, W., He, X., Liu, P., Zhang, C., Zhao, X., Liu, C., Ma, Z., Liu, J., Wang, J., Lu, K., Catoire, V., Mellouki, A. and Mu, Y.: Atmospheric Measurements at the Foot and the Summit of Mt. Tai – Part II: HONO Budget and Radical (RO_x + NO_3) Chemistry in the Lower Boundary Layer, Atmos. Chem. Phys. Discuss., 2021, 1–36, doi:10.5194/acp-2021-531, 2021.

Ye, C., Xue, C., Zhang, C., Ma, Z., Liu, P., Zhang, Y., Liu, C., Zhao, X., Zhang, W., He, X., Song, Y., Liu, J., Wang, W., Sui, B., Cui, R., Yang, X., Mei, R., Chen, J. and Mu, Y.: Atmospheric Hydrogen Peroxide (H_2O_2) at the Foot and Summit of Mt. Tai: Variations, Sources and Sinks, and Implications for Ozone Formation Chemistry, J. Geophys. Res. Atmos., 126(15), 1–14, doi:10.1029/2020JD033975, 2021.

2. Response to RC2

This paper presents comprehensive field campaign which was performed in summer at the foot (150 m a.s.l) and the summit (1534 m a.s.l) of Mt. Tai (Shandong province, China). The author performed the analysis of HONO budget and found strong unknown HONO sources. Constraints on the kinetics of aerosol-derived HONO sources were discussed and their contribution to HONO formation were negligible. The vertical transport form the ground to the summit levels and heterogeneous conversion of NO_2 was proposed to support the remaining majority of unknown HONO sources. The subject is suitable for publication in ACP and I would recommend the paper is accepted after the author have addressed the following concerns.

Response: Thanks for your efforts and comments, which help to improve our manuscript. Please see the point-to-point response below (**Comments in Black; Response in Blue; Changes in Red**).

Specific comments:

Instrumentation: Low levels of HONO were measured by LOPAP technique with detection limit of 1.5 pptv at the summit of Mt. Tai in summer 2018. The QA and/or QC for LOPAP instrument should be stated to guarantee data quality.

Response: In Section 2.2 we added more information about the operation of the LOPAP instrument. At the summit station, a temperature-controlled measurement container was used to house all the instruments. The external sampling unit of LOPAP was installed on the top of the container, about 2.5 m above the ground surface. Zero air (ultrapure N_2) measurements were conducted 2 or 3 times per day. Liquid calibration with diluted standard nitrite solution (Sigma-Aldrich) was conducted every week. Both zero air measurements and liquid calibration were conducted after changing any solution, cleaning the instrument, or replacing any component of the instrument (the air pump was broken on 21st July and replaced by a new one on 25th July). The precision of the instrument determined from 2σ noise of the calibration was 1%. An accuracy of 7% was determined by error propagation including all known uncertainties, i.e., the concentration of the calibration standard (±3-4%) and the liquid (±1%) and gas flow (±2%) rates. Known artificial HONO formation on inlet surfaces (e.g., Zhou et al., 2002) were minimized by using the external sampling unit, with only a 3 cm sunlight-shielded glass inlet to the ambient atmosphere. Other interferences were considered of minor importance, as they were corrected for by the two-channel concept of the instrument. In addition, excellent agreement between LOPAP and DOAS techniques was observed under complex conditions in a smog chamber and in the ambient atmosphere (Kleffmann et al., 2006).

Anthropogenic emissions: The author stated that low NO_x/NO_y of 0.43 ± 0.28 indicated aged air masses and small impact of anthropogenic emissions. However, NO and NO_x were measured simultaneously at the summit station. Why did not the author utilize NO/NO_x to evaluate the influence of nearby anthropogenic emissions. Moreover, the rapid increase in pollutants (HONO, NO, NO_2, NOy, CO, $PM_{2.5}$) was observed on 29 July. Low NO concentrations (1-2 ppbv) were observed at high O_3 levels (~50 ppbv), which should originate from local emissions. However, the author stated the high HONO levels could come from the heterogeneous conversion. The author should reexamine the data and explore the sources of increased pollutants.

Response: As demonstrated in Section 2.1, potential anthropogenic emissions could happen around the Southern Heavenly Gate, the Bixia Temple, and the Jade Emperor Peak. All of the three places are within 1 km west of our station. If emissions originated from those regions, sharp peaks would be observed and the NO/NO_x ratio should be near to that of fresh plumes.

However, this event lasted about 1.5 hours (5:20-6:50), much longer than the duration of the fresh plumes observed at the foot station. Besides, during this event, air mass originated from the south (Figure 2), the polluted urban region rather than the direction of the potential sources at the summit level. Furthermore, the NO/NO_x ratio in this plume is 0.21, lower than fresh combustion plume with a NO/NO_x ratio of ~0.9 or even higher (Carslaw and Beevers, 2005; He et al., 2020; Kurtenbach et al., 2012; Wild et al., 2017). This is also lower than the fresh plumes observed at the foot station with an average NO/NO_x ratio of 0.46±0.19 at high O_3 levels (Xue et al., 2021).

Therefore, we could conclude that the observed plume should originate from transport from the foot urban region rather than nearby emissions at the summit.

We then improved related texts as:

During this event, air mass originated from the south (Figure 2), the polluted urban region (Figure S1E) rather than the direction of the potential sources at the summit level. This event lasted about 1.5 hours (5:20-6:50), much longer than the duration of the typical fresh plumes observed at the foot station. Furthermore, the NO/NO_x ratio of this plume was 0.21, lower than the direct NO/NO_x emission ratio of ~0.9 (Carslaw and Beevers, 2005; He et al., 2020; Kurtenbach et al., 2012; Wild et al., 2017). This is also lower than that of the fresh plumes observed at the high-O_3 foot station with an average NO/NO_x ratio of 0.46±0.19 (Xue et al., 2021). Therefore, we could conclude that the observed plume should originate from the foot urban region rather than nearby emissions at the summit. The $\Delta HONO/\Delta NO_x$ within this plume was 8%, much larger than that inferred from direct emissions (typically inferred as less than 1%). The ratio could be enhanced by: 1) night-time NO_2-to-HONO conversion at the ground level where the air mass was already aged before being transported to the summit level, 2) in-plume NO_2-to-HONO conversion along the mountain slope (rock and vegetation surfaces, etc.), and 3) in-plume NO_2-to-HONO conversion on particle surfaces as both the boundary layer height (BLH) elevation and the valley breeze are initialized after sunrise.

Figure 5: The data of HONO and $J(NO_2)$ for summit and foot station were measured at different periods. Whether it is appropriate to exhibit the data at different periods together in the figure? The measured data at different periods were different. Is such comparison meaningful?

Response: Measurements at the foot and the summit stations represent typical average diurnal variations for ground surface or summit measurements, respectively. Similar pattern of variations have also been reported by many previous studies including ground surface measurements (Alicke et al., 2002, 2003; Gu et al., 2020; Hendrick et al., 2014; Kleffmann et al., 2005; Platt et al., 1980; Su et al., 2008) and summit measurements (Jiang et al., 2020; Kleffmann et al., 2002; Kleffmann and Wiesen, 2008). Besides, our measurements at the two stations were conducted during two consecutive periods in summer 2018. To confirm our argument, we also compared pollutants at the ground and summit stations during the same period, such as $PM_{2.5}$, CO, O_3, and SO_2 (Figure 7 in the manuscript) discussed in Section 3.2.2.3 of our manuscript.

Hence, the comparison could allow potential insights into the link between atmospheric chemistry at the ground surface and summit levels.

Page 13, line 285-290: The author stated that south wind could enhance the upslope valley breeze wind because higher wind speeds (>5 m s^{-1}) were observed at the summit station than at the foot of the mountain (> 2 m s^{-1}). However, the wind speeds are generally higher at the summit station, which requires detailed explanation by the author.

Response: The fact that the south wind could enhance the upslope valley breeze wind is not because of higher wind speed at the summit level. It's because the urban site (150 m a.s.l.) is south of the summit station (1534 m a.s.l.).

The reported upslope valley breeze wind speed was about 2-5 m s^{-1}. With consideration of south wind at the ground level (>2 m s^{-1}), the integrated wind speed along the mountain slope could be 4-7 m s^{-1}. Alternatively, with consideration of south wind at the ground level (>5 m s^{-1}), the integrated wind speed along the mountain slope could be 7-10 m s^{-1}. Therefore, we used the wind speed range of 4-10 m s^{-1} to consider all the possible situations.

Related texts are improved as:

The upslope valley breeze wind could transport polluted air mass from the foot to the summit levels. This process could be accelerated by the dominant south wind (Figure 8) as the urban site (150 m a.s.l.) is south of the summit station (1534 m a.s.l.). The mean south winds measured at the ground and summit stations are >2 and >5 m s^{-1}, respectively. Then the integrated wind speed along the mountain slope should be 4 – 10 m s^{-1}, and the calculated $t_{transport}$ will be reduced to 7 – 17.5 min.

Page 16, line 347-348: "Note that the uncertainty of …". I don't quite understand this sentence. Section 3.6 stated the contribution of photolysis of HONO and O_3 to OH. Please give the explanation.

Response: We cited Figure 10 and added the below discussion on the contribution (3%) of $P(HONO)_a$ to HONO formation. It has been revised as:

Note that the uncertainty of S_a is not expected to cause a significant uncertainty on HONO budget analysis as $P(HONO)_a$ was not the dominant source (Figure 10 and see the below discussion on $P(HONO)_a$ contribution).

The author calculated the enhanced uptake coefficient of NO_2 on the aerosol surfaces. The dark uptake of NO_2 on the aerosol surface could be considered to evaluate the influence of heterogeneous reaction on the aerosol surfaces since the dark uptake coefficient of NO_2 were mostly investigated.

Response: From the correlation analysis (Table 4), we found poor correlations (r = 0.17 or 0.64) between P_{un} and NO_2*S_a or $NO_2*S_a*J(NO_2)$, suggesting minor roles of dark and photo-enhanced NO_2 uptake on the aerosol surface in the HONO formation.

Besides, with $\gamma_a = 2\times10^{-5}$, photo-enhanced $P(HONO)_a$ could only explain 3% of P_{un}. For dark NO_2 uptake, γ_{a_dark} is generally at a level of 10^{-6} (George et al., 2005; Han et al., 2017; Stemmler et al., 2006, 2007), implying that $P(HONO)_{a\_dark}$ is much lower than $P(HONO)_a$. Therefore, we didn't consider dark NO_2 uptake on the aerosol surface.

The following sentence was added in Section 3.4:

Note that dark NO_2 uptake on the aerosol surface was not considered due to a much lower uptake coefficient generally at a level of 10^{-6} (George et al., 2005; Han et al., 2017; Stemmler et al., 2006, 2007).

Page 19, Section 3.6: The author only calculated the contribution of the photolysis of HONO and O_3 to OH and not HO_x. The HO_x should be replaced by OH.

Response: Done.

Page 21, line 470: What dose λ_a stand for? It is γ_a?

Response: It has been changed to γ_a.

Reference

Alicke, B., Platt, U. and Stutz, J.: Impact of nitrous acid photolysis on the total hydroxyl radical budget during the Limitation of Oxidant Production/Pianura Padana Produzione di Ozono study in Milan, J. Geophys. Res. Atmos., 107(D22), 8196, doi:10.1029/2000JD000075, 2002.

Alicke, B., Geyer, A., Hofzumahaus, A., Holland, F., Konrad, S., Pätz, H.-W., Schäfer, J., Stutz, J., Volz-Thomas, A. and Platt, U.: OH formation by HONO photolysis during the BERLIOZ experiment, J. Geophys. Res. Atmos., 108(D4), 8247, doi:10.1029/2001JD000579, 2003.

Carslaw, D. C. and Beevers, S. D.: Estimations of road vehicle primary NO_2 exhaust emission fractions using monitoring data in London, Atmos. Environ., 39(1), 167–177, doi:10.1016/j.atmosenv.2004.08.053, 2005.

George, C., Strekowski, R. S., Kleffmann, J., Stemmler, K. and Ammann, M.: Photoenhanced uptake of gaseous NO_2 on solid organic compounds: A photochemical source of HONO?, Faraday Discuss., 130, 195–210, doi:10.1039/b417888m, 2005.

Gu, R., Zheng, P., Chen, T., Dong, C., Wang, Y., Liu, Y., Liu, Y., Luo, Y., Han, G., Wang, X., Zhou, X., Wang, T., Wang, W. and Xue, L.: Atmospheric nitrous acid (HONO) at a rural coastal site in North China: Seasonal variations and effects of biomass burning, Atmos. Environ., 229, 117429, doi:10.1016/j.atmosenv.2020.117429, 2020.

Han, C., Yang, W., Yang, H. and Xue, X.: Enhanced photochemical conversion of NO_2 to HONO on humic acids in the presence of benzophenone, Environ. Pollut., 231(1), 979–986, doi:10.1016/j.envpol.2017.08.107, 2017.

He, L., Zhang, S., Hu, J., Li, Z., Zheng, X., Cao, Y., Xu, G., Yan, M. and Wu, Y.: On-road emission measurements of reactive nitrogen compounds from heavy-duty diesel trucks in China, Environ. Pollut., 262, 114280, doi:10.1016/j.envpol.2020.114280, 2020.

Hendrick, F., Müller, J.-F., Clémer, K., Wang, P., De Mazière, M., Fayt, C., Gielen, C., Hermans, C., Ma, J. Z., Pinardi, G., Stavrakou, T., Vlemmix, T. and Van Roozendael, M.: Four years of ground-based MAX-DOAS observations of HONO and NO_2 in the Beijing area, Atmos. Chem. Phys., 14(2), 765–781, doi:10.5194/acp-14-765-2014, 2014.

Jiang, Y., Xue, L., Gu, R., Jia, M., Zhang, Y., Wen, L., Zheng, P., Chen, T., Li, H., Shan, Y., Zhao, Y., Guo, Z., Bi, Y., Liu, H., Ding, A., Zhang, Q. and Wang, W.: Sources of nitrous acid (HONO) in the upper boundary layer and lower free troposphere of the North China Plain: insights from the Mount Tai Observatory, Atmos. Chem. Phys., 20(20), 12115–12131, doi:10.5194/acp-20-12115-2020, 2020.

Kleffmann, J. and Wiesen, P.: Technical Note: Quantification of interferences of wet chemical HONO LOPAP measurements under simulated polar conditions, Atmos. Chem. Phys., 8(22), 6813–6822, doi:10.5194/acp-8-6813-2008, 2008.

Kleffmann, J., Heland, J., Kurtenbach, R., Lörzer, J. and Wiesen, P.: A New Instrument (LOPAP) for the Detection of Nitrous Acid (HONO), Environ. Sci. Pollut. Res., 9(special issue 4), 48–54, 2002.

Kleffmann, J., Gavriloaiei, T., Hofzumahaus, A., Holland, F., Koppmann, R., Rupp, L., Schlosser, E., Siese, M. and Wahner, A.: Daytime formation of nitrous acid: A major source of OH radicals in a forest, Geophys. Res. Lett., 32(5), L05818, doi:10.1029/2005GL022524, 2005.

Kleffmann, J., Lörzer, J. C., Wiesen, P., Kern, C., Trick, S., Volkamer, R., Rodenas, M. and Wirtz, K.: Intercomparison of the DOAS and LOPAP techniques for the detection of nitrous acid (HONO), Atmos. Environ., 40(20), 3640–3652, doi:10.1016/j.atmosenv.2006.03.027, 2006.

Kurtenbach, R., Kleffmann, J., Niedojadlo, A. and Wiesen, P.: Primary NO_2 emissions and their impact on air quality in traffic environments in Germany, Environ. Sci. Eur., 24(21), 1–8, doi:10.1186/2190-4715-24-21, 2012.

Platt, U., Perner, D., Harris, G. W., Winer, A. M. and Pitts Jr, J. N.: Observations of nitrous acid in an urban atmosphere by differential optical absorption, Nature, 285(5763), 312–314, doi:10.1038/285312a0, 1980.

Stemmler, K., Ammann, M., Donders, C., Kleffmann, J. and George, C.: Photosensitized reduction of nitrogen dioxide on humic acid as a source of nitrous acid, Nature, 440(7081), 195–198, doi:10.1038/nature04603, 2006.

Stemmler, K., Ndour, M., Elshorbany, Y., Kleffmann, J., D'Anna, B., George, C., Bohn, B. and Ammann, M.: Light induced conversion of nitrogen dioxide into nitrous acid on submicron humic acid aerosol, Atmos. Chem. Phys., 7(16), 4237–4248, doi:10.5194/acp-7-4237-2007, 2007.

Su, H., Cheng, Y. F., Shao, M., Gao, D. F., Yu, Z. Y., Zeng, L. M., Slanina, J., Zhang, Y. H. and Wiedensohler, A.: Nitrous acid (HONO) and its daytime sources at a rural site during the 2004 PRIDE-PRD experiment in China, J. Geophys. Res. Atmos., 113(D14), D14312, doi:10.1029/2007JD009060, 2008.

Wild, R. J., Dubé, W. P., Aikin, K. C., Eilerman, S. J., Neuman, J. A., Peischl, J., Ryerson, T. B. and Brown, S. S.: On-road measurements of vehicle NO_2/NO_x emission ratios in Denver, Colorado, USA, Atmos. Environ., 148, 182–189, doi:10.1016/j.atmosenv.2016.10.039, 2017.

Xue, C., Ye, C., Kleffmann, J., Zhang, W., He, X., Liu, P., Zhang, C., Zhao, X., Liu, C., Ma, Z., Liu, J., Wang, J., Lu, K., Catoire, V., Mellouki, A. and Mu, Y.: Atmospheric Measurements at the Foot and the Summit of Mt. Tai – Part II: HONO Budget and Radical (RO_x + NO_3) Chemistry in the Lower Boundary Layer, Atmos. Chem. Phys. Discuss., 2021, 1–36, doi:10.5194/acp-2021-531, 2021.

Zhou, X., He, Y., Huang, G., Thornberry, T. D., Carroll, M. A. and Bertman, S. B.: Photochemical production of nitrous acid on glass sample manifold surface, Geophys. Res. Lett., 29(14), 26-1-26–4, doi:10.1029/2002GL015080, 2002.