Comments to the author:

Dear Authors,

This time, the revised manuscript as well as the Authors' responses were evaluated by the Reviewer #1 and myself. Though several parts were improved, still major revision of the manuscript is needed. See further comments from the reviewer #1 and my points below.

Response: Thanks for the comments from the editor and reviewer #1. Please see the point-to-point response below (Comments in Black; Response in Blue; Changes in Red). Line numbers mentioned in this reply refer to our revised version with changes tracked.

1. About estimated OH: the authors' description was improved but the main point from the reviewer #1 is still not addressed. It is read from line 347 (change-track version) that relationship between OH and J(O1D) was taken from (Kanaya et al., 2009) for the summit of Mt. Tai for June 2006 but it is not found there. Is this relationship from other study? Clarification is necessary. Irrespective of the answer to this question, OH concentration levels need to be estimated "with" the measured HONO concentration in this case for consistency with other parts of this study. One possibility would be to linearly scale the OH levels with the enhancement of the primary production due to the HONO photolysis, while a full box model simulation (i.e., deriving HO2 levels and quantifying OH production from the HO2 + NO reaction) is recommended. I believe that the P_un cannot be explained by this revised treatment but consistency and quantitativeness are necessary. Also please note that OH from Kanaya et al. (2009, 2013) was not MEASURED but "estimated" from a box model constrained with observations (lines 351-352).

Response: Regarding the relationship of OH and J(O^1D), it was indeed taken from Kanaya et al. (2009), in which radical chemistry was modeled and discussed. From the averaged diurnal variations of J(O^1D) (Fig. 1) and the modeled OH (Fig. 2) high correlations (R^2>0.9) were found between them, indicating very similar diurnal variations of OH to that of J(O^1D). However, this does not yield high correlations between the time series of modeled OH and that of J(O^1D), as the editor commented that correlation between OH and J(O^1D) was not found during the MTX 2006 campaign. Therefore, we can conclude that the present OH estimation method can predict the diurnal variations of OH but not its absolute levels due to 1) high correlations between OH and J(O^1D) was found in their average diurnal variations but not in their time series, and 2) HONO was not constrained in the box model simulations for the MTX 2006 campaign.

One way to consider HONO impact is to discuss the uncertainties caused by HONO when using the correlation between OH and J(O^1D) during the MTX 2006 campaign. Assuming that the impact of HONO on OH levels is determined by its contribution to primary OH production at both the foot and the summit stations, we can preliminarily deduce the present OH estimation uncertainties caused by the lack of HONO chemistry. At the foot station, HONO photolysis made a contribution of 64% to primary OH production (see Section 3.6). If the box model was not constrained by the measured HONO, OH would be underestimated by 25% (see the companion paper, Xue et al.

(2022)). At the summit station, HONO contributed 18% of the primary OH production (see Section 3.6). Therefore, OH underestimation due to HONO chemistry at the summit station should be roughly around 7% (25% divided by 64% and multiplied by 18%).

Regarding the suggestion on box model simulations, because VOCs data were not available, we can't run box models with VOCs and HONO constrained to estimate OH levels.

Considering that Mt. Tai is surrounded by polluted regions, empirical formulas between OH and $J(O^1D)$ from the ground or other mountain measurements, may not be reasonable here. Hence, to cover the uncertainties caused by the above issues, we made sensitivity tests on OH levels by increasing or reducing its level by 30%.

As the editor/reviewer also agrees with our argument that the OH level doesn't significantly impact P_{un} as well as further analysis based on the obtained P_{un}, we will improve the text about how the present OH was estimated and point out its uncertainties as well.

Changes in the manuscript (L346-373):

OH measurements were not available during this campaign. One popular used method to estimate OH is based on the correlation between OH and solar ultraviolet radiation (e.g., $J(O^1D)$) (Rohrer and Berresheim, 2006). Considering that Mt. Tai is surrounded by polluted regions, empirical formulas between OH and $J(O^1D)$ from the ground or other mountain measurements, may not be reasonable here. In June 2006 Kanaya et al. conducted a comprehensive field campaign at the summit of Mt.Tai (Kanaya et al., 2009). OH levels and sources were studied by a box model. From the average diurnal variations of the modeled OH and $J(O^1D)$, a significant correlation ($R^2>0.9$) between them was found, which was used here to estimate OH concentrations. This method could lead to some uncertainties in OH levels due to: 1) high correlations between OH and $J(O^1D)$ was found in their average diurnal variations but may not be in their time series, and 2) HONO was not constrained in the box model simulations so that OH could be underestimated for the MTX 2006 campaign.

One way to consider HONO impact is to discuss the OH uncertainties caused by the lack of HONO chemistry. Assuming that the impact of HONO on OH levels is determined by its contribution to primary OH production, we can preliminarily deduce the OH uncertainties at the summit station caused by the lack of HONO chemistry based on measurements and model simulations for the foot station. At the foot station, HONO photolysis made a contribution of 64% to primary OH production (see Section 3.6). If the box model was not constrained by the measured HONO, OH would be underestimated by 25% (see the companion paper, Xue et al. (2022)). At the summit station, HONO contributed 18% of the primary OH production (see Section 3.6). Therefore, OH underestimation due to the lack of HONO chemistry at the summit station should be roughly around 7% or so.

Hence, to cover the uncertainties caused by the above issues, we added OH sensitivity tests, reducing or increasing the OH level by 30%, to quantify the impact of the OH uncertainties in our further 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 modeled during the MTX campaign (Kanaya et al., 2009, 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%). Reducing or enlarging OH levels by 30% 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 small impact of variable OH and $HONO_{pss}$ levels on P_{un}. Hence, we highlight the uncertainties in OH levels estimated by the current method, but its impact on following HONO budget analysis should be small as discussed above.

2. The concept that HONO is transported with the upslope wind is interesting, but is valid if only after verifying that dispersion/diffusion during the transport is also slower than the time constant. This dispersion/diffusion term could be estimated from longer-lived species (e.g., CO or SO2). Such discussion is needed before conclusion in lines 329-333.

Response: Thanks for the suggestion. Discussion about dilution and dispersion is a very good point to have more insights into the concept proposed in this study. We compared CO levels (thanks for the guidance) at the foot and the summit stations to preliminarily deduce the dilution effect and revised the related discussion.

(L320-330)

Figure 9 shows the calculated α with $t_{transport}$ = 7 or 17.5 min during the daytime. It is apparent that α is larger than 43% with $t_{transport}$ = 17.5 min and larger than 72% with $t_{transport}$ = 7 min, providing a theoretical basis for the potential role of vertical HONO transport from the ground to the summit levels. The calculations don't consider the atmospheric dilution or dispersion during the transport, which may reduce α. This effect could be roughly quantified by comparing levels of long lifetime species (e.g., CO) at the foot and the summit stations. Hourly CO averages at noon are 493 and 379 ppbv measured at the foot and the summit stations, respectively (Figure 5). This preliminarily indicates a dilution factor of 1.3. The dilution process may also similarly affect HONO, i.e., α is expected to be reduced by a factor of 1.3, leading to α values of >55% and >33% with $t_{transport}$ = 7 or 17.5 min, respectively. The above calculation only included the daytime HONO sink through photolysis and atmospheric dilution, but the sources, such as NO + OH and heterogeneous NO_2 reactions, were not considered, and hence, the calculated α represents a lower limit.

3. OH generation from H2O2 photolysis would be of minor importance (4% of OH production term from Kanaya et al, 2009, or even less when considering HONO photolysis) and thus the description in lines 445-450 is not readily supported. The authors need a quantitative counter statement about the contribution when the statement is to be kept.

Response: Thanks for the information about the contribution of H_2O_2 photolysis to OH. We agree that the contribution indeed is expected to be less when HONO was considered.

Besides, we estimated the OH production from H_2O_2 photolysis ($P(OH)_{H2O2}$). With noontime H_2O_2 of 1.4 ppbv measured in summer 2019 (Ye et al., 2021) and maximum photolysis frequency of 7.0×10^{-6} s^{-1}, the calculated $P(OH)_{H2O2}$ is 0.04 ppbv h^{-1}, which is much lower than $P(OH)_{HONO\_net}$ or $P(OH)_{O3}$ of 0.2 and 1.0 ppbv h^{-1}, respectively. Therefore, we deleted the discussion about the role of H_2O_2 in OH production.

4. The righthand side of the equation 8 must be [HONO]xJ(HONO) - k_1[NO][OH] - k_2[HONO][OH]. Here the OH levels are again necessary to take into account (similar to the point 1 above; the OH level should be determined with the impact from HONO photolysis). Correction is necessary when the calculations were not made adequately.

Response: The typing mistake in Equation 8 was corrected. Note that during our calculation in Excel, the right formulas were used.

Regarding the comment on OH level, please see the above response to comment 1.

References

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