Response to Reviewer 3:

The work by Jiang et al presents measurements of HONO and other supporting species at Mt Tai, a mountaintop site in the North China Plain. Concurrent ground level measurements were also performed, and comparison were made to explore the source differences. The authors present an interesting data set and provide modelling work to aid their interpretation. Overall, this paper is well thought out and written, with the results clearly presented in the tables and figures. I would recommend publication after consideration to the comments below.

Response: We appreciate the reviewer for the positive comments and helpful suggestions. We have carefully considered all of the comments, and revised the original manuscript accordingly. Below we list the original referee's comments in black *italics*, followed by our responses and changes in the manuscript shown in blue and red, respectively.

Minor comments

1. Section 2.2: Were the same instruments used at Mt. Tai as the ground level monitoring stations? This is particularly important for HONO measurements, as previous work has shown significant differences can be reported for co-located HONO instruments, even of the same type (See e.g. Crilley et al., 2019). Furthermore, were there any inter-comparison measurements of the instruments at Mt Tai and ground to account for any differences between instruments that may affect the comparison?

Response: We are sorry that the original description may be misleading. The same LOPAP instrument was used at both Mt. Tai and the ground-level site (Ji'nan). Please note that both measurement campaigns were not concurrent, and the 1-yr continuous HONO measurements in Ji'nan were carried out from September 2015 to August 2016, earlier than the present study at Mt. Tai (2017 winter and 2018 spring).The measurement protocol and quality assurance and quality control procedures of LOPAP were also the same between the two measurement campaigns. For clarity, the following statements have been provided in the revised manuscript.

"It should be noted that a 1-yr continuous measurement campaign had been conducted from September 2015 to August 2016 at an urban site of Ji'nan using the the same instrument (Li et al., 2018a), and their results are compared here with those at Mt. Tai to infer the vertical distributions of HONO in the NCP region."

2. Page 5, line 3: Typically, the baseline is measured every 4-8 hours with a LOPAP (see e.g. Crilley et al., 2019; Kleffmann and Weisen 2008) to capture the temporal variability. Measuring the baseline every 11h 30 min may not be sufficient to capture the baseline variability and I am curious why the authors chose to do it like this.

Response: Thanks for the helpful suggestion. Considering the relatively clean conditions at such a high elevation station, we chose to perform the baseline correction in a relatively longer time interval (i.e., 11 h 30 min) in this study. We further examined the temporal variability of the measured baseline during this study, and it was quite stable (see figure below). We believe that this choice should not affect the quality of observations presented here, and will take this into consideration in our future studies with different pollution conditions.

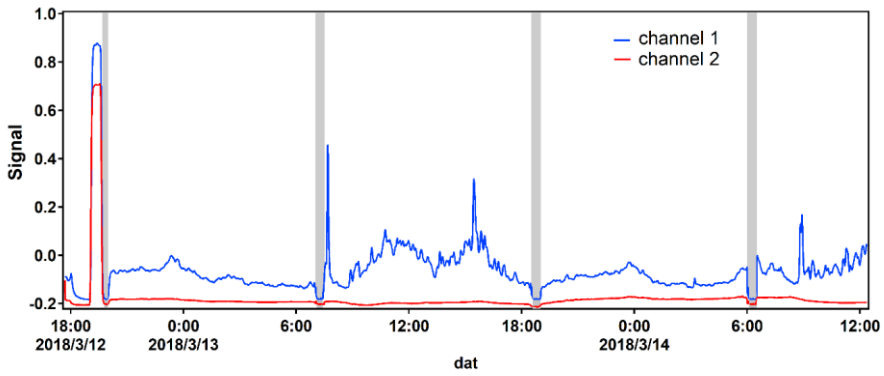

Figure R3-1. Raw signal of LOPAP instrument showing the baseline calibration (grey shaded).

3. Page 6, line 22: I don't quite follow this sentence 'based on the CO (temperature) data and the measured correlations with CO (temperature) for anthropogenic (biogenic) VOCs' What do you mean by CO (temperature)?

Response: We are sorry that the original descriptions may be too simplified and unclear. We have elaborated more about it in the revised manuscript by the following discussions.

"For VOCs and carbonyls for which the measurements were not in real-time, the high-resolution data were approximated as follows. During the daytime when multiple VOC and carbonyl samples were available, the measurement data were interpolated to a time resolution of 5 min. For the period when measurement data were unavailable, the VOC concentrations (except for isoprene) were estimated with the real-time CO data by assuming a linear regression relationship between VOCs and CO (note that the regression was established with the available measurement data of VOCs and CO). The same method was applied for isoprene and carbonyls, but ambient temperature was used instead of CO for isoprene, and multi-linear regression with CO and O_3 was used for carbonyls to account for the primary and secondary sources of carbonyls (Yang et al., 2018; Xue et al., 2016). Such approximation may be subject to some uncertainties but should not significantly interfere the estimation of the role of HONO photolysis in OH sources (Yang et al., 2018)."

4. Page 10, line 23: are the reported j(HONO) and OH concentrations noontime maxima or daily average?

Response: They are the average values at noontime (11:00-14:00 LT). The original statement has been revised as follows for clarity.

"According to the measurement-derived J(HONO) (with noontime averages of 6.4±3.5 and $9.5\pm3.2\times10^{-4}$ s^{-1} in winter and spring; see Fig. S3) and the model-simulated OH concentrations (with noontime averages of 2.5±0.7 and $4.4\pm2.0\times10^{6}$ molecules cm^{-3}; Fig. S3), the average lifetime of HONO was estimated as 25.7±1.4 and 21.8±16.9 minutes during noontime (11:00-14:00 LT) in winter and spring, respectively."

5. Page12, line7: Here you state that heterogenous reaction with NO_2 on aerosol surfaces should be a significant daytime HONO source at Mt. Tai rather than on ground. What is ratio of ground vs aerosol surface area? I am asking to try and understand why this NO_2 reaction may preferentially occur on aerosol surfaces at Mt Tai, unlike previous work at ground level.

Response: Actually, we cannot make an accurate estimation for the ratio of ground versus aerosol surface areas. Here we argue that aerosol surface may play a more important role than ground surface in the heterogeneous formation of HONO mainly due to the following reasons. First, Mt. Tai (1534 m a.s.l.) is the highest mountain over the North China Plain, and the station is situated on an isolated peak of the mountain. Thus, the measurement site is far away from the ground level surface, and the terrestrial surface is much limited compared to the ground level studies. Second, the correlation analysis showed much stronger correlation between P_{other} and NO_2*(S/V)a (the commonly used indicator for the heterogenous HONO formation on aerosol surface) than that between P_{other} and NO_2 (the indicator for heterogeneous HONO formation on ground surface). More measurements are still needed to better understand the potential role of aerosol surface in the HONO formation in the high elevation atmospheres.

6. Page 13, line 10: it would also be good to report the percentage HONO photolysis contributes to OH production from the model, as this would enable comparison to other work.

Response: Thanks for the helpful suggestion. The following statements have been added in the revised manuscript.

"In percentage, HONO photolysis accounted for 44.4% and 25.8% of the total primary RO_x production at mid-day at Mt. Tai in winter and spring, respectively. For OH alone, the percentages of the contribution of HONO photolysis to the primary sources were 93.2% and 71.3% in winter and spring."

7. Figure 3: Why is there so much more noise in the HONO/NO_2 diurnal plots compared to HONO and NO_2?

Response: We examined the measured time series of HONO, NO_2 and HONO/NO_2 ratios, and indeed found larger fluctuations in the HONO/NO_2 ratio than those of HONO and NO_2. The fluctuation in HONO/NO_2 was amplified by the variability of NO_2 concentrations, with much higher HONO/NO_2 values at lower NO_2 levels. So, the large fluctuations in HONO/NO_2 should be partly due to the relatively lower levels of NO_2 at Mt. Tai. In comparison with the ground-level site in urban Ji'nan, the fluctuation in the measured HONO/NO_2 was much smaller given its much higher levels of NO_2.

8. Figure 5: I am surprised that there is no noon-time maxima in $HONO_{pss}$, as the OH should peak then (as seen in Fig 8) along with the NO_x? (as shown in Fig 2)?

Response: We are sorry that Figure 5 with a log coordinate for y-axis was not clear to show the noon-maxima in $HONO_{pss}$. As shown from the figure below, HONOpss actually showed a noontime maximum, and the diurnal pattern of $HONO_{pss}$ followed well with that of [NO]*[OH].

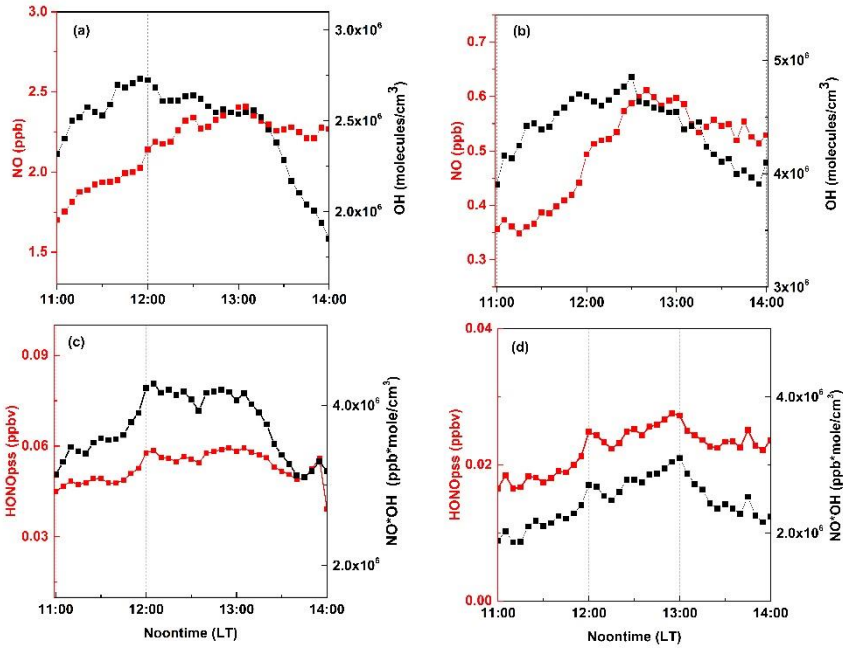

Figure R3-2. Average mixing ratios of NO, OH, $HONO_{pss}$ and [NO]*[OH] concentrations around noon (11:00-14:00 LT) in winter (a and c) and spring (b and d).