Response to the Review Comments

We thank the reviewers for providing insightful comments and helpful suggestions that have substantially improved the manuscript. Below we have included the review comments in italic followed by our responses in blue. In the revised manuscript, we have highlighted those changes accordingly with changes tracked. The line number refers to the ones in the tracked change version manuscript.

Reviewer #1:

The authors present a measurement-based study to report high wintertime HONO concentrations in Nanjing in the YRD area in China, and investigate the contribution of HONO to the atmospheric oxidation capacity in terms of its contribution to the OH formation using a box model. They attribute the high HONO level to the NO2 heterogeneous reactions on surfaces (particles and ground). Although some results are surprising (for example, photolysis of HONO is the dominant daytime OH source in this area), the methods are overall sound, the paper is well structured and well written in general. The manuscript can be accepted for publication after addressing the following minor issues.

One of the major concerns is that the authors rely considerably on the correlation analysis to reach conclusions, but some of the correlation information may not be valid (a good correlation between two variables does not necessarily mean a cause-and-effect relationship between them). For example, the authors use "The observed similarity between HONO/NO2 and HONO in diurnal profiles" to "strongly suggests that HONO in the study area was likely originated from NO2 heterogeneous reactions". The similarity between HONO/NO2 and HONO merely suggests that NO2 does not importantly affect the HONO diurnal variation or NO2 concentrations are relatively time-invariant (which is indeed the case as shown in Fig. 3). This similarity does not provide any connections between the HONO formation and the NO2 heterogeneous reaction. Another questionable example is that the authors use "concurred elevated HONO and PM2.5 levels" to "strongly suggest that high HONO may increase the atmospheric oxidation capacity." Although there is no doubt that high HONO would enhance the atmospheric oxidation capacity and therefore SOA formation, but one could also argue that elevated PM2.5 levels lead to elevated HONO due to the heterogeneous reactions and elevated PM2.5 levels could be due to emissions. Therefore, the only information of concurrence of elevated HONO and PM2.5 levels may not necessarily suggest enhanced oxidation capacity.

Response: We agree with the reviewer that good correlation does not necessarily mean a-cause-and-effect relationship between the two variables. The statement in lines 40-41 has been revised as: "Model simulations indicated that heterogeneous chemistry played an important role in HONO formation".

Indeed, we cannot quantify how much of the $PM_{2.5}$ was actually from secondary formation. Therefore, we have softened the statement in lines 48-51. It has been revised as "Our study indicated that elevated $PM_{2.5}$ level during the haze events can promote NO_2 to HONO conversion by providing more heterogeneous reaction sites and hence increase the atmospheric oxidation capacity, which may further promote the formation of secondary air pollutants".

Specific comments:

(1) In Eq. 9, the authors appear to attribute the term of Punknown to the heterogeneous NOx reactions (including photosensitized and non-photosensitized). In fact, this term should also include HONO emissions and transport (advection). This may partially explain the moderate correlation coefficients (~0.5) between this term and (NO2).NO2.RH or J(NO2).NO2.S/V.RH. Although the authors claim that HONO emissions are negligible, the OH production results from two industrial plumes in Fig 6 could also suggest the importance of HONO emissions (besides NO and VOC emissions) to HONO and OH.

Response: We have included the primary emission term in Eqs. 9 and 10 to evaluate its contribution to the total budget of HONO. Both Figs. 9 and 10 have been revised to include the emission term.

$$\frac{d[HONO]}{dt} = (P_{OH+NO} + P_{emission} + P_{unknown}) - (L_{OH+HONO} + L_{photolysis} + L_{deposition}) \quad (9)$$

$$P_{unknown} = \frac{d[HONO]}{dt} + L_{OH+HONO} + L_{photolysis} + L_{deposition} - P_{OH+NO} - P_{emissions}$$

$$= \frac{d[HONO]}{dt} + k_{OH+HONO}[OH][HONO] + J_{HONO}[HONO] + \frac{v_{HONO}}{H}[HONO]$$

$$-k_{OH+NO}[OH][NO] - \frac{0.003\Delta NOx}{\Delta t} \quad (10)$$

Although primary emission of HONO was not substantial but it indeed should not be neglected. In this work, we treated the study area as a standalone "box" and the impact of transport (advection) was not considered. All reactive chemicals affecting HONO were assumed in a steady state and the box model was constrained by the measured species, including VOCs, NO_x, HONO, J-values, and other trace gases. The impact of PM loading on HONO production was also evaluated using the box model in the revised manuscript as suggested by the other reviewer.

(2) In the box model, how is the time variation of the PBL height considered? How it is represented may affect the agreements between observed and simulated HONO concentrations in Fig 10. The authors may also discuss how other limitations or assumptions in the box model affect the simulation results.

Response: The PBL height was based on the remote sounding measurements conducted in Nanjing, a station maintained by the Institute for the Environment (IENV) at the Hong Kong University of Science and Technology (HKUST) (http://envf.ust.hk/dataview/profile/current/). The diurnal PBL

profile used here was hourly averaged of all measured data points within December 2015. The uncertainty associated with the model simulations including the limitations in PBL and other input variables were assessed by Monte Carlo analyses. In each Monte Carlo simulation, the input variables of the model, including HONO, O_3, NO, NO_2, CO, SO_2, HCHO, VOCs, reaction rate constants, PBL height, and photolysis frequencies, were independently set to vary within ±10% of the mean value of individual variable with a normal probability distribution (see Lines 222-227). Overall, the Monte Carlo sensitivity analysis show that the model uncertainty of HONO ranged from ±13% to ±38% (see Fig. 10a for details). The sensitivity analysis reinforced the conclusions that the proposed heterogeneous sources can generally capture the observed HONO trend (see Lines 545-549). Also inserted into Fig. 10a are the contributions of primary HONO emission denoted by the brown color. Indeed, primary emission was not trivial. However, primary emission evidently did not contribute significantly to the total HONO budget.

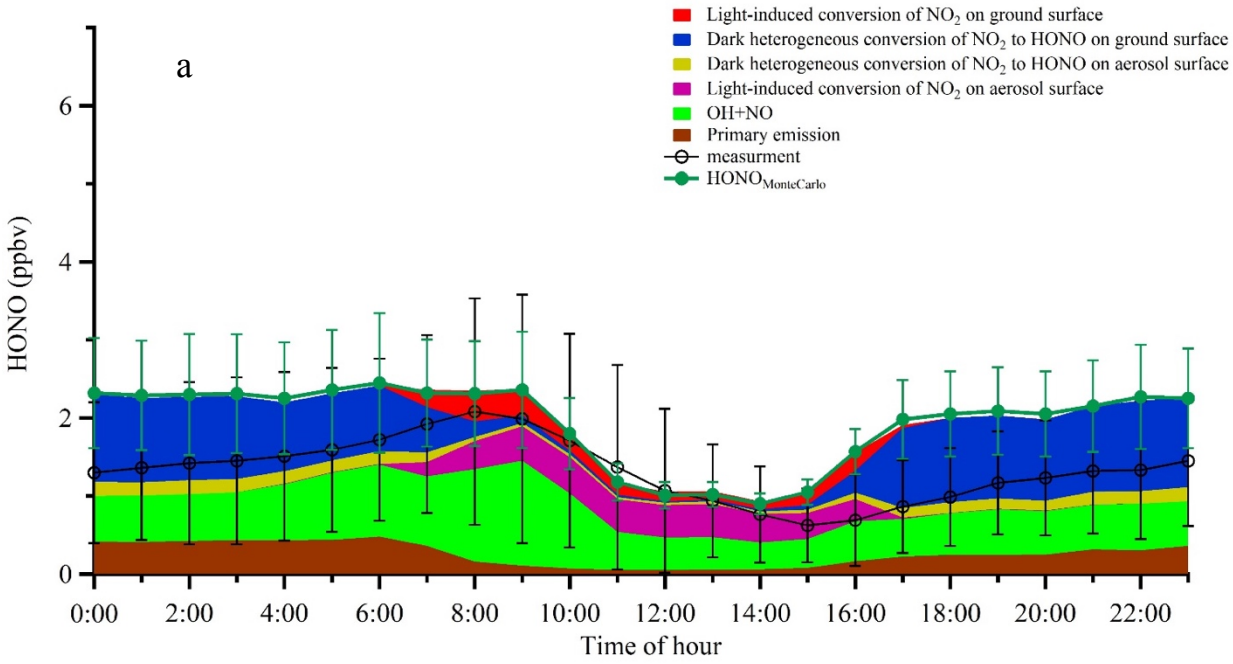

Figure 10a. Averaged diurnal profiles of the measured HONO and the modeled HONO from different sources. Error bars on the black line represent standard deviations of HONO measurements in hourly bins. Error bars on the green markers denote the Monte Carlo analysis results.

Technical corrections:

(1) When several numbers in the same units stand site by site, it is better to just use unit once (e.g., L240, 266, 297-298, 515-517).

Response: Those repeated units have been removed.

(2) In eqn 9, should ∂[HONO] be d[HONO]? ∂t dt

Response: The Eqs. 9 and 10 all derivatives have been revised into "d/d..." and primary emission is also considered in the equations. Please see response to specific comment #1.

(3) Fig 9, Photolysis should be Lphotolysis.

Response: Figure 9 legend of photolysis loss has been revised into "$L_{photolysis}$".

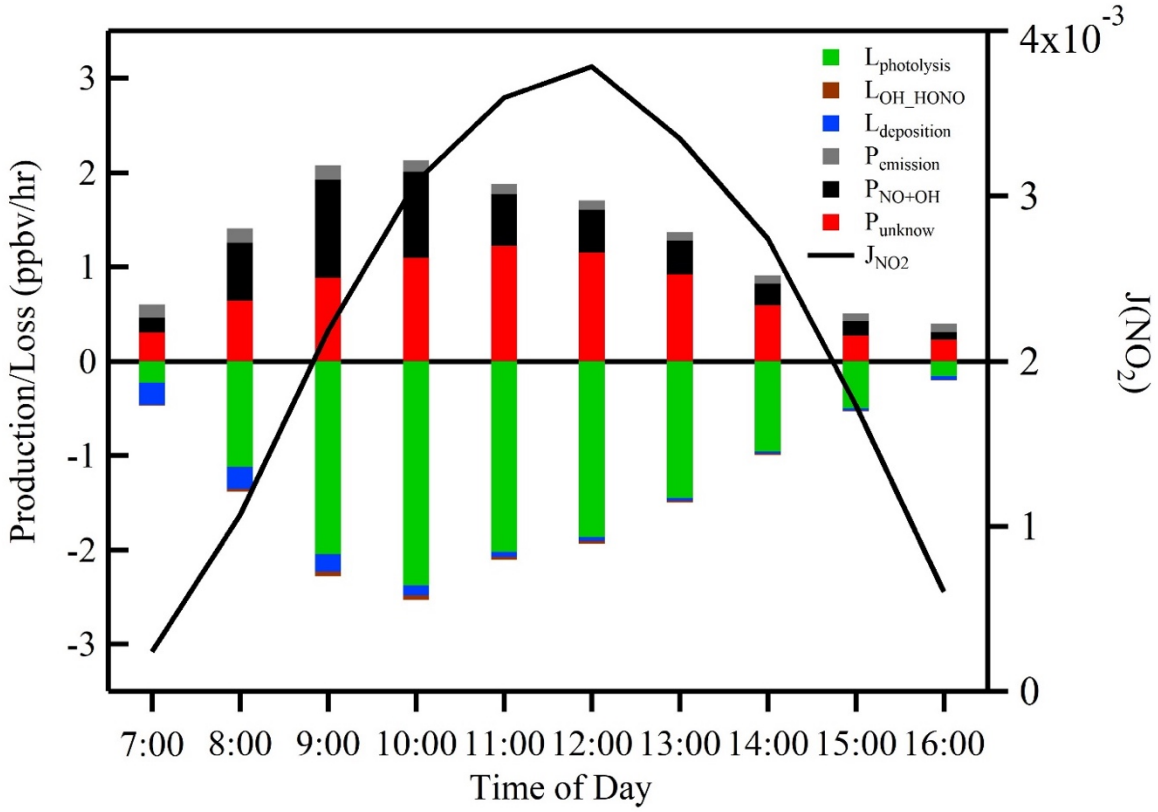

Figure 9. Averaged production and loss rates of daytime HONO and J(NO_2) during the measurement period. The black line shows the photolysis rate of NO_2.

Reviewer #2:

This work combines comprehensive field measurements and full box model simulation to investigate the unknown sources of HONO and the contribution to atmospheric oxidation capacity. The results and conclusions highlight the large contribution of HONO to OH radical source and the important role of light-induced enhanced heterogeneous processes in daytime HONO formation. Overall, the manuscript is written with clear logic, fluent language, deep analysis and full discussion. However, there are some major and minor comments which require to be addressed before the manuscript is accepted.

Major comments:

1. With consideration of the high concentrations of NO (dozens of ppbv) from time to time in the morning from Fig. 2, suggest carefully calculating and assessing the contribution of primary emissions from urban vehicle exhausts to the HONO source.

Response: As suggested by the reviewer, we have conducted additional model simulations including primary emission as one of the sources. The simulation results are shown in Fig. 10a. Indeed, HONO from primary emissions was not negligible and the model simulations show that it accounts for 11% of the total HONO concentration. We have included the primary emission term in Eqs. (9) and (10). The uncertainty of the model simulation results was assessed by Monte Carlo analysis and the results are displayed in Fig. 10a as green error bars. Overall, the simulation uncertainty varied from ±13% to ±38%.

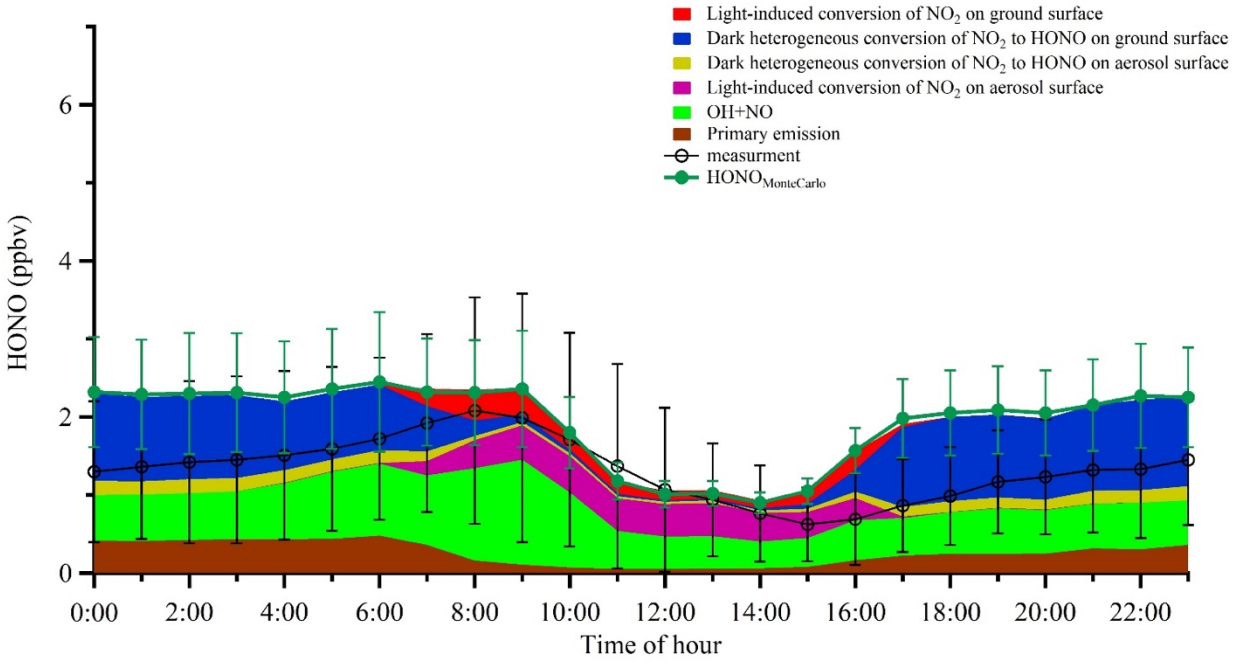

Figure 10a. Averaged diurnal profiles of the measured HONO and the modeled HONO from different sources. Error bars on the black line represent standard deviations of HONO measurements in hourly bins. Error bars on the green markers denote the Monte Carlo analysis results.

2. Light-induced enhancement of HONO formation on aerosol surfaces was found by the authors, particularly within industrial plumes. So, what photosensitized aerosol components were

responsible for the photo-catalyzed processes? Aerosol composition data or additional discussions are required to elaborate this issue.

Response: We agree with the reviewer it would be ideal to have aerosol chemical composition information to support our conclusion. Unfortunately, aerosol chemical composition was not measured during this work. However, since the photo-enhanced HONO formation only proceeds on aerosol surfaces, aerosol chemical composition may not be able to directly reflect aerosol surface chemical property. The detailed mechanisms underlying the light-induced HONO formation on aerosol surfaces are still under active research. It is believed to be highly dependent on the surface properties. For example, the conversion rates on soot surface (Han et al., 2017) and organic surface (Stemmler et al., 2006) can vary substantially. In addition, laboratory studies have found that this mechanism is not catalytic by the surface in nature and the rate of which may vary with the availability and aging state of the surface reaction sites. Therefore, aerosol chemical composition alone may not be sufficient to deduce the actual HONO formation mechanism. Currently, in atmospheric chemistry models the light-induced HONO formation process was only treated as a bulk reaction on the aerosol surface with a substantial faster surface uptake coefficient of NO_2 (Huang et al., 2017; Li et al., 2018; Wang et al., 2017). We have added the following statement in the manuscript (Lines 482-489) to further elaborate this issue.

"Since aerosol chemical composition was not measured in this work, we cannot demonstrate any possible direct connection between aerosol composition and the photo-enhanced HONO formation on aerosol surfaces. Nevertheless, the actual mechanism underlying the photo-enhanced HONO formation on aerosol surface need further investigation. It has been found that photo-sensitized NO_2 conversion rate coefficient on different surfaces can vary substantially (Han et al., 2017; Stemmler et al., 2006). Furthermore, studies have shown that this type of surface reaction is not catalytic in nature and the surface reaction rate may vary with the availability and aging state of the surface reaction sites (Stemmler et al., 2006). Therefore, aerosol chemical composition alone may not be sufficient to reveal the actual HONO formation processes."

3. The accurate simulation by using one-dimension box model requires stable meteorological conditions. In this study, how to select the simulation periods and what are the criteria?

Response: The model we used in this study is a box model or 0-dimensional (not one-dimensional) model which does not require stable meteorological conditions. The box model was constrained by the observed meteorological conditions (temperature, pressure, humidity, photolysis frequencies) and chemical species. We agree with the reviewer that meteorological conditions can significantly affect the air pollution level at an observation site through transport or convection. A 3-D model would be ideal to investigate both physical and chemical processes that could affect the HONO chemistry. However, the accuracy of a 3-D model would rely heavily on the accuracy of emission inventory and meteorological field used in the model, which were basically unavailable for the study area. In addition, to accomplish a 3-D modeling work was far beyond the scope of this work.

The objective of this work was to investigate the impacts of HONO chemistry on the local atmospheric oxidative capacity through a comprehensive field campaign at one supersite. Since no transport and vertical mixing were considered here, a 0-D box model should be sufficient to do the job. Especially the observation period was during winter time, when wind field was relatively

weak with an average wind speed of 1.7 m s^{-1} and the solar radiation was less intense, leading to weaker vertical mixing. Therefore, the winter weather condition would make the observation site prone to local air pollution accumulation, which would justify the usage of a simple 0-D box model.

Minor comments:

1. Line 37–39, point out the detailed period of time.

Response: These numbers are campaign averaged results. The exact campaign period "from 1 to 31 December 2015" has been inserted into the sentence in Line 32 after "China". A phrase "during the campaign period" has been inserted in Line 37 after "The results show that...".

2. Line 172, how about the zero calibration or background detection for the HONO measurement? With zero air or using other method?

Response: As described in Section 2.1 and illustrated in Fig. 1, a configuration of two samplers in serial is used with the first sample to measure the total signal and the second sampler to measure background signal. The difference between the two samplers is the net HONO signal. The background signal is usually only a few percent of the total signal. We have added the following few sentences (Lines 165-168):

"Two coil samplers in serial were used to measure total signals in the first sampler and the background in the second sampler. The difference between the two samplers is the net HONO signal. The background signal is usually only a few percent of the total signal."

The instrument calibration was carried out by injecting standard $NaNO_2$ solution into the instrument right after the sampling coil. During the campaign period, the instrument was calibrated once every four days and the HONO data in between calibrations were adjusted with the calibration factors accordingly.

We have inserted the following statement into the supporting information to further explain the HONO instrument operation: (Lines 184-186)

"The instrument calibration was carried out once every four days by injecting standard sodium nitrite ($NaNO_2$) solution into the instrument right after the sampling coil.

3. Line 206-207, describe the potential uncertainty aroused by the limited VOC species used in the box model.

Response: In this work, a GC-FID instrument was used to measure 60 VOCs, including most alkanes, alkenes, and aromatics. Although oxygenated VOCs (OVOCs) were missing except formaldehyde and a few other carbonyls detected by a DHPL method, we constrained the box model with measured VOCs. The model results show that OVOCs only accounted for a very small portion of the total VOCs in this industrial area. Moreover, OVOCs even contributed much less to the total VOC OH reactivity. Therefore, we believe the missing OVOC (except carbonyl compounds) would not significantly affect our model simulation results. We have inserted the following statement into the manuscript (Lines 222-227):

"Although the oxygenated VOCs (OVOCs) other than formaldehyde and some other carbonyls (by the DNPH method) were not measured in this study, they were simulated in the box model that

was constrained to measured VOC. Our results indicated that OVOCs only accounted for a small portion of the total VOCs in this industrial area and even contributed much less to the total VOC OH reactivity. Therefore, the limited VOCs detected in this work would not significantly affect the following model simulation results."

4. Line 214, "VOC" can be "VOCs".

Response: It has been changed to VOCs.

5. Line 235, 238, 239, suggest adding a word such as "averaging" before the "maxiuma" and "minimum".

Response: We have inserted "daily averaged" before "maxima" and "minimum".

6. Line 241–243, the "similarity between the diurnal profile of HONO/NO2 ratio and that of HONO" cannot suggest that HONO was likely originated from NO2 heterogeneous reactions. Unchanged NO2 concentration also led to similar trends between HONO/NO2 ratio and HONO concentration. Similar trends between HONO/NO2 ratio and S/V ratio could be an evidence for heterogeneous formation on aerosol surfaces.

Response: We agree with the reviewer and the statement has been removed to avoid misleading.

7. Line 295, state the possible uncertainty or influence on the model simulation results due to the assumption of constant H2O2 concentration of 3 ppbv. A linear or non-linear estimation of H2O2 concentration from other pollutants or parameters is better than a constant value.

Response: The H_2O_2 level is unknown in the study area and H_2O_2 measurements were seldom conducted in China. However, H_2O_2 is much less photo-sensitive than other major OH precursors, such as O_3, HCHO and HONO. Model simulation tests with doubled H_2O_2 concentration can only cause a few more percent increase in OH production from H_2O_2 photolysis, which was significantly lower than the model uncertainty estimated by the Monte Carlo analysis. Therefore, we believe H_2O_2 would not contribute significantly to the total OH budget estimation. Moreover, estimation of H_2O_2 as a function of other parameters cannot be supported by our other measurements that may also introduce other much larger uncertainties.

8. Line 300, point out the detailed period of time, e.g., 7:00-16:00 local time.

Response: We have modified the sentence in Line 321

"As shown in Fig. 5, the contribution of HONO photolysis to OH production during 7:00-16:00 local time varied from 23.6% to 63.3% with a mean value of 44.8%."

9. Line 334–336, are there PM2.5 composition data to support that most of the PM2.5 components came from secondary formation during the two industrial plume events and the enhancement of secondary aerosol components simultaneously occurred with elevated HONO photolysis rate? If

there is no enough evidence, suggest removing the deduction that "high levels of HONO promote the formation of PM2.5" in the main text, Abstract, and Highlights.

Response: $PM_{2.5}$ composition was not measured in this work. The statement in the abstract has been removed. The sentence in Line 356 has been revised as:

"Although ambient OH concentrations during these events may not be high (see Fig. 4a), the high levels of HONO can boost active photochemical oxidation and thus promote the formation of other secondary air pollutants."

Highlight#3 has been revised as:
"High loading of $PM_{2.5}$ provided additional reaction surfaces for HONO formation."

10. Line 342, the HONO emission ratio was expressed as HONO/NOx ratio, right?

Response: Yes, the HONO emission ratio was expressed as HONO/NOx ratio. It has been revised accordingly.

11. Line 350– 351, with consideration of the high concentration of NO particularly in the morning (see Fig. 3), the influence of traffic source possibly cannot be ignored. Suggest carefully evaluating the influence from traffic source.

Response: As suggested by both reviewers we have included the primary HONO emission into the model simulation to assess the impacts of primary emission more accurately. We have revised Section 3.5.1 :

"Previous studies have demonstrated that HONO can be emitted directly from vehicle exhaust (Kirchstetter et al., 1996; Kurtenbach et al., 2001). However, the NO/NOx ratio measured in this work was relatively low, with an average of 0.25±0.06, much less than that of freshly emitted exhausts (> 0.9) obtained from tunnel experiments (Kurtenbach et al., 2001), indicating that the air masses sampled in this work had been considerably aged and mixed with other air masses, and hence primary HONO from direct emission (if there was any) had been diluted substantially (less than a few per cents) before reaching the observation site. In addition, our sampling site is located nearby the industrial zone, and the high concentration of NOx was mainly originated from the industrial activities, so the influence of traffic source on HONO was expected to be small. To further evaluate the potential impact of primary emissions on HONO concentration, we have incorporated the contribution of primary HONO emission into the MCM box model. The HONO emission ratios, i.e., HONO/NOx, was taken as 0.3% (Kirchstetter et al., 1996), representing a gasoline-fueled vehicle fleet, which was very typically encountered in the study area. On average, the primary emissions from vehicle exhaust can only account for 11% of the total HONO concentration, indicating secondary mechanisms still dominated HONO level in the study area, which will be further analyzed in the following sections."

In addition, the simulation results are included in the new Fig. 10a. Please see response to the specific comment #2 of reviewer #1 for details. Indeed, although primary HONO emission was not substantial, it should not be neglected.

12. Line 357, as to "the two different time points", how were the time periods selected? Based on what criteria?

Response: There is no commonly-accepted criteria for time period selection that is used to calculate the NO_2 to HONO conversion ratio. In summary, we searched through the HONO and NO_2 timeseries for periods when both of them increased monotonically with a correlation coefficient higher than 0.8. The following sentence has been inserted into Line 390-392:

"The time periods used to calculate HONO/NO_2 conversion ratio were selected when both HONO and NO_2 increased monotonically with a correlation coefficient higher than 0.8."

13. Line 374, as to "for several individual days", which days? What are the criteria for the selection?

Response: The individual days referred to days when industrial plumes were encountered at the site, i.e., the 7, 21, and 22 of December. The data on these days have been used to generate Fig. 10b. In addition, we have conducted additional model sensitivity study with respect to aerosol surface area. Therefore, we have removed these correlation analysis results from the manuscript.

14. Line 397, "timely" can be temporal.

Response: "Timely" has been changed to "temporal".

15. Line 413, carefully evaluate the HONO emission from traffic sources, due to sometimes NO concentration was very high, particularly in the morning (see Fig. 3).

Response: The primary HONO emission has been included in the daytime HONO budget evaluation. We have revised the sentence as: "The impact of HONO direct emissions was relatively small at daytime." (Line 456)

16. Line 498–500, were there aerosol composition data to show the fractions of secondary components and primary components during the industrial plume events? Were there high levels of photosensitized components such as metals, black carbons, and brown carbons?

Response: Aerosol chemical composition was not measured during this work. We have downplayed the discussions on the correlations between HONO and $PM_{2.5}$. Please see responses to major comments #2 and minor comments #9.

17. Line 524–525 and the subsequent paragraph, was the enhanced HONO formation during daytime was dominated by humid heterogeneous reactions, or photosensitized reactions, or together?

Response: New model simulation including HONO primary emission showed that the enhanced HONO formation during daytime was mainly due to photosensitized reactions on both aerosol surface (28.2%) and ground surface (17.8%). The heterogeneous mechanism only accounted for 2.2% from aerosol surface and 7.9% from ground surface. The sentence in Lines 583-587:

“The model suggests that higher daytime levels of HONO were mainly produced by the light-induced conversion of NO_2 on aerosol surfaces (28.2%) and ground surfaces (17.8%) (except early morning). While the heterogeneous HONO production on ground surface dominated nocturnal HONO sources, heterogeneous reactions on various surfaces only contributed a small portion of total HONO at daytime (2.2% on aerosol surface and 7.9% on ground surface).”

Reviewer #3:

1) This manuscript reports the results of a field campaign in Nanjing, a megacity within the Yangtze River Delta (YRD) region, during December, 2015. HONO and related species were simultaneously measured. High levels of (especially daytime) HONO were reported and were most likely due to heterogeneous reactions involving NO2. YRD is one of the most developed and also polluted regions of China. Atmospheric oxidation capacity (mostly determined by the OH radical) is the fundamental driving force that is responsible for the fast formation of secondary air pollutants such as O3 and PM. Although observations of high levels of HONO are not surprising in China, such a comprehensive field campaign like this study is still of practical importance to fully understand the role of HONO chemistry in this region. Especially, the budget of OH radical in the YRD region shall be extensively assessed. The subject of this study is within the scope of ACP. Overall, the experimental methodology is generally sound and the measurements were properly conducted. The manuscript is fairly well written and the logic is clear to follow.
One of my major concerns is that the authors claim that primary emission did not contribute significantly to the observed HONO. But I would suggest the authors to further evaluate the significance of primary emission using the box model, which should be able to give a more reliable and quantitative assessment of primary emitted HONO.

Response: We appreciate the helpful suggestion from the reviewer and we have conducted additional model simulations including primary emission as one of the sources. The emission ratios of HONO with respect to NO_x ($HONO/NO_x$) was taken from the results obtained from tunnel measurements of Kirchstetter et al. (1996), i.e., 0.3%, which represented a gasoline powered vehicle fleet, which was typically encountered in the study area. Also, it should be noted that some the measured HONO may be originated from heterogeneous reactions on various surfaces during the tunnel experiments. Therefore, the estimated HONO emissions should be taken as an upper limit. The simulation results are shown in Fig. 10a. Indeed, although the contribution from primary HONO emission was relatively small, it should not be neglected.

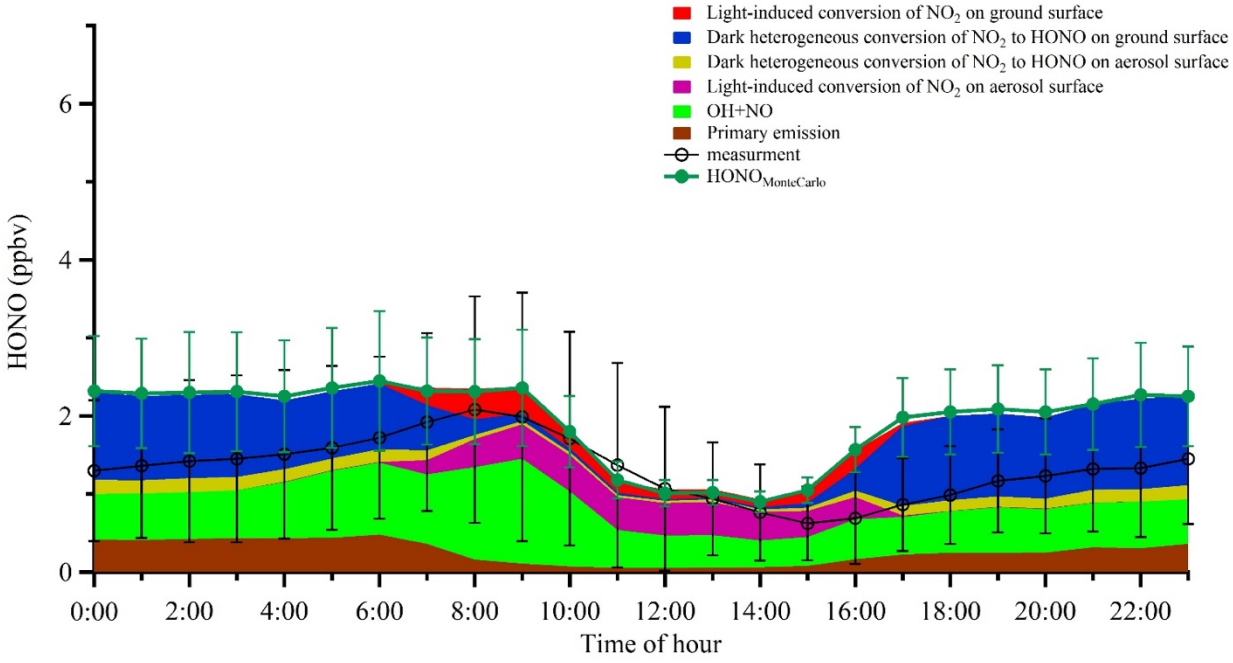

Figure 10a. Averaged diurnal profiles of the measured HONO and the modeled HONO from different sources. Error bars on the black line represent standard deviations of HONO measurements in hourly bins. Error bars on the green markers denote the Monte Carlo analysis results.

2) Also, I would suggest the authors to weaken the role of correlation analysis (Section 3.5.5) and rely more on the model simulation results. The good correlation between aerosol surface density and HONO does not necessarily mean that HONO is produced on aerosol surfaces. A model sensitivity study would be a better way to verify if high loading of PM was playing an important role in HONO formation by promoting heterogeneous reactions.

Response: Yes, we agree with the reviewer that a simple correlation analysis is not sufficient to establish the connection between HONO formation and aerosol surface area. We have conducted extra model sensitivity study to verify the impacts of aerosol surface on HONO production. By decreasing and increasing the aerosol surface area density by a factor of 2, respectively. The results showed that the contribution from heterogeneous photosensitized conversion of NO_2 on aerosol surfaces would correspondingly vary from 18% to 40% of the total HONO budget. Therefore, we believe aerosol surface played an important role in HONO photosensitized formation in the study area. A statement has been inserted into Lines 545-549:
"In addition, we have performed a model sensitivity study with respect to aerosol surface density by varying S/V from 50% to 200% of the average value. The results showed that the contribution from heterogeneous photosensitized conversion of NO_2 on aerosol surfaces would correspondingly vary from 18% to 40% of the total HONO budget, demonstrating that aerosol surface chemistry played an important role during HONO formation in the study area."

Technical comments:
1) L103: "because"
2) L273: "heterogeneous"
3) L329: "the origins of these..."
4) L368: "appears"

Response: The above typos have been corrected and the manuscript has been revised accordingly.

References:
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