Referee #1

This paper reports year round measurements of HONO in Nanjing , Eastern China. The effect of direct emissions of HONO is calculated by looking at fresh plumes and production of HONO from heterogeneous reaction of NO2 on aerosol surfaces was speculated upon using nighttime HONO and RH measurements. A HONO budget was calculated, along with a missing HONO source. The effect of the measured HONO on the OH budget was also described. Understanding the role of HONO is crucial for the understanding of oxidation chemistry, especially in the urban environment therefore this study is important work that should be published. There are very few long term measurements of HONO in the literature, with most studies being done in short term campaigns. The analysis here is a reasonable attempt at understanding the role of HONO, albeit with a fairly limited set of supporting measurements. It is within scope of ACP and I recommend publication subject to completion of the following modifications.

General comments:

-No OH measurements were available during the measurements period so the authors have calculated OH concentrations for their analysis (P7). They use the work of Rohrer and Berresheim that correlates OH with J(O1D). I find this a strange choice of literature to use as it was based on work in a very different environment. I believe there are numerous measurements of OH taken within the PRD that would be a more relevant way to infer OH concentrations for this study.

Response: Thanks for the comment and suggestion. We have reset the parameters a, b, and c, based on the OH studies in the PRD, China.

Line 190-201: The coefficient a reflects the general chemical conditions (e.g. NO_x or VOCs) at the selected place for research, and the exponent b represents the combined effects of all photolytic processes on OH, and the parameter c counts the light-independent OH sources. The values of a and b in Eq. (2) are adopted from the study in the Pearl River Delta (Lu et al., 2012). The value of c is set to 1.0×10^{6} cm^{-3}, a typical nighttime OH concentration in urban areas of China (Li et al., 2012;Lu et al., 2014). The calculated OH concentrations around noon were in the range of $0.15\text{-}1.6\times10^{7}$ cm^{-3}, comparable to observations in Chinese urban atmospheres (Lu et al., 2012;Lu et al., 2013).

$$[OH]=a\times(J(O^{1}D)/10^{-5}s^{-1})^{b}+c \\ (a=5.6\times10^{6}cm^{-3},\ b=0.68,\ c=1.0\times10^{6}cm^{-3}) \quad (2)$$

-The authors present a calculation of the effect of HONO on OH formation and compare it

to formation by O3 photolysis. This study needs expanding a little. OH production from the HO2 + NO reaction would likely be the largest source in such an environment as this study. If the authors want to look at HOx radical formation then they should also make some comment about the effect of other sources such as HCHO photolysis to form HO2 and O3 + alkene reactions. I realise they may not have the supporting measurements to do this accurately but some mention should be made of it.

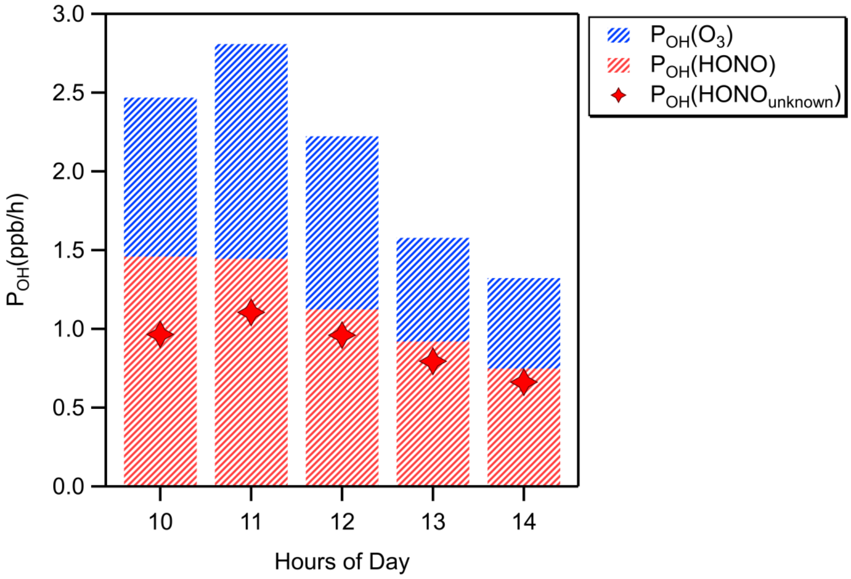
Response: Thanks for the comment and suggestion. We have added the content about the sources of OH radicals in the revised manuscript.

Line 279-285: In addition to the two mechanisms mentioned above, there are other pathways to generate primary OH radicals: the photolysis of aldehydes, especially HCHO, can form HO_2 *radicals, and then react with NO to form OH radicals; the reaction of ozone with alkenes produce OH radicals directly; the ozonolysis of alkenes and nighttime reactions of* NO_3 *radicals with alkenes can also be net sources of OH radicals (Finlayson-Pitts and Pitts, 2000;Seinfeld and Pandis, 2016).*

-Some comment should be made as to how much the 'missing HONO' source contributes to OH. This is important in terms of understanding how much models might be underestimating OH by not having all the HONO sources in them.

Response: Thanks for the comment and suggestion. We have quantified the relative contribution of the missing HONO source to OH in the revised manuscript.

Line 549-552: In our study, the OH production rate from the missing HONO accounts for about 78% of total $P_{OH}(HONO)$ *(Fig. S2), suggesting that the unconventional source of HONO is of significance to atmospheric oxidation.*



Average OH production rates from photolysis of HONO, the missing HONO and O_3 around noon (10:00-14:00 LT), from Nov. 2017 to Nov. 2018.

-Following on from this, some mention should be made about how various air quliaty forecasting or regional models treat HOHO and how adding in the 'unconventional' sources might affect oxidation chemistry.

Response: Thanks for the comment and suggestion. We have recommended a possible mechanism that can be adopted into the models in the in the revised manuscript, i.e. the photo-induced heterogeneous reaction of NO_2.

Line 572-577: Our study suggest that the missing source of HONO should be considered in the air quality forecasting or regional models to characterize atmospheric oxidizing capacity better, especially in warm seasons (spring and summer). Based on the measurement (Fig. S3), the light-induced heterogeneous conversion of NO_2 *to HONO on aerosol surfaces and ground surface can been included in simulation works probably, as what did in the study of Lee et al. (2016).*

Minor comments:

-P5 line 150: Can the authors justify that the measurement is 'interference free'? It was my understanding that the LOPAP instrument is subject to interference from other nitrate species. Please expand this.

Response: Thanks for the comment and suggestion. The interferences can be reduced mostly but possibly not completely by subtracting the signal of channel 2 from the signal of channel 1, so we have modified the statement in the revised manuscript

Line 143-146: In the first stripping coil, all of the HONO and a fraction of interfering substances were absorbed into solution, and the remaining interfering species (NO_2, HNO_3, *PAN, etc.) were absorbed in the second stripping coil.*

Line 149-151: The real HONO signal was the difference between the signals in the two channels, and the interferences can be minimized by this method.

P8 line 244: 'in the' should be 'at'

Response: Thanks for the suggestion. We have corrected this in the revised manuscript.

Line 248-251: Given that the photolytic lifetime of HONO is about 10-20 min at the midday (Stutz et al., 2000), the considerable HONO concentration during daytime indicates the existence of strong production of HONO.

P9 Line 264: The authors describe HONO as an efficient reservoir of OH radicals. I'm not totally convinced this is the correct way to describe it. A reservoir suggests a long lived species that enables transport of OH radicals. I would have thought the lifetime of HONO would be very short, maybe the authors could comment on this.

Response: Thanks for the comment and suggestion. We have modified the statements in the revised manuscript.

Line 14-16: Nitrous acid (HONO), a important precursor of the hydroxyl radical (OH), has been long-standing recognized to be of significance to atmospheric chemistry, but its sources are still debate.

Line 269-270: The elevated mixing ratio of HONO presents an efficient source of OH radicals during daytime in Nanjing.

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Referee #3

This manuscript describes continuous ambient measurements of HONO, NO, NO2, and PM2.5 at the SORPES station in Nanjing (eastern China) from November 2017 to November 2018. The main conclusions are:

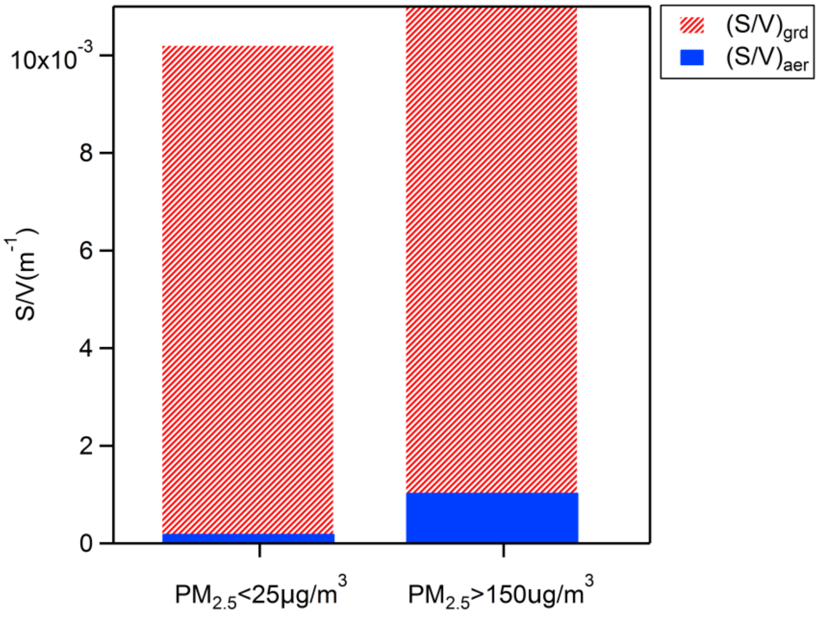
1. Seasonal average HONO concentrations are comparable to other urban/suburban regions (0.45-1.04 ppb). 2. Direct emissions from combustion sources explain nearly 25% of nocturnal HONO concentrations. The authors determined this by examining fresh plumes. 3. Nocturnal HONO formation is RH-dependent and largely explained by heterogeneous surface chemistry. 4. A missing diurnal HONO formation mechanism is a significant source of HONO around noon (average 1.13 ppb/hr).

There are not many long-term records of ambient HONO measurements, and this manuscript provides a valuable dataset to the scientific community. It is well within the scope of ACP and will likely be of interest to ACP readers. I recommend publication after the authors address the following comments.

-Figure 6 shows the HONO/NO2 ratio as a function of RH. The authors state that Fig 6a represents measurements when available surface area is dominated by the ground (i.e., relatively low surface area contributions from aerosols). Can the authors quantify the relative contributions to total surface area from the ground and aerosols? What percentage of the total surface area does the ground represent in clean air and polluted air?

Response: Thanks for the comment and suggestion.

We calculated aerosol surface density from the particle number size distributions between 6 nm and 820 nm, by assuming that all particles are spherically shaped. and We calculated ground surface density through the equation: $(\frac{S}{V})_{grd} = \frac{1}{H}$, where H is the height of boundary layer, and a value of 100m is assumed for nighttime (Su et al., 2008). As the following figure shows, the surface area to volume ratio of ground is dominant, but under the condition of severe pollution, the aerosol can contribute about 10% of the total surface area. Besides the surface area, the conversion of NO_2 to HONO should also be determined by the surface reactivity, i.e. the uptake coefficient of NO_2-to-HONO ($\gamma_{NO_2 \rightarrow HONO}$). Differ from the prolonged exposure to oxidizing agents and radiation of the ground surface, the aerosol surface is relatively more fresh, and possibly more reactive. For example, the reduction of NO_2 in the presence of water by C–O and C–H groups in the soot is proposed to produce HONO quickly (Ammann et al., 1998). In our study, in case we assume that all of the observed HONO is formed on particle surfaces at night, the derived $\gamma_{NO_2 \rightarrow HONO}$ is 1.44×10^{-5}, within a reasonable range of laboratory measurements.



the averaged surface area to volume ratio (m^{-1}) of ground and aerosol in clean air ($PM_{2.5}$<25μg/m^3) and polluted air ($PM_{2.5}$>150μg/m^3)

Line 481-487: For 30%-100% of the measured mean C_{HONO} *(0.0043* h^{-1}*) in winter, the uptake coefficient of* NO_2*-to-HONO* ($\gamma_{NO_2 \rightarrow HONO}$) *calculated from Eq. (8) is in the range of* 6.9×10^{-6} *to* 1.44×10^{-5}*, consistent with the results from many laboratory studies which demonstrate that the uptake coefficients of* NO_2 (γ_{NO_2}) *on multiple aerosol surfaces or wet surfaces are mainly distributed around* 10^{-5} *with the HONO yield varying from 0.1 to 0.9 (Grassian, 2002;Aubin and Abbatt, 2007;Khalizov et al., 2010;Han et al., 2017).*

-As shown in Figure 6, HONO/NO2 ratios in polluted air do not decline at RH between 75-95% as is seen in clean air. The authors should provide some explanation here. Why is there a different RH dependency under high PM2.5 conditions?

Response: Thanks for the comment and suggestion. We have added some discussion into the revised manuscript.

Line 427-438: With the increase of RH, the hygroscopic growth of aerosol particles should provide larger surface area. When RH is higher than 75%, which has exceeded the mutual deliquescence relative humidity of inorganic salts (Fountoukis and Nenes, 2007), aerosols will transfer to aqueous phase gradually, and then promoting multiphase or heterogeneous chemistry processes (Herrmann et al., 2015). For example, the oxidation of SO_2 *by* NO_2 *on aqueous aerosol surface may produce* NO_2^-*/HONO efficiently under polluted condition (Xie et al., 2015;Wang et al., 2016). In addtion, the enhancement* NO_2 *uptake on micro-droplets by anions has been reported in experiments (Yabushita et al., 2009).*

-The authors claim that the unknown daytime HONO production is different from the heterogeneous nocturnal production (section 3.4). It is not immediately clear how the authors reach this conclusion. They should expand on this statement and provide clear justification.

Response: Thanks for the comment and suggestion. The highest noontime $P_{unknown}$ value is 1.72 ppb/h in spring, followed by 1.11 ppb/h in summer, 0.66 ppb/h in autumn and 0.58 ppb/h in autumn, unlike the seasonal variation of NO_2; and $P_{unknown}$ shows an increase towards noon, which is also distinguished from the diurnal pattern of NO_2. These results indicate that there must be some other factors affecting $P_{unknown}$, in case NO_2 is assumed to be a dominate precursor of HONO at daytime

Line 558-561: The average value of $P_{unknown}$ normalized by NO_2 is 0.1 h^{-1}, over 18 times greater than the nighttime conversion rate (0.0055 h^{-1}), also implying that $P_{unknown}$ cannot be explained by the nocturnal mechanism of NO_2-to-HONO.

-A major justification for assuming an unknown HONO source is that the HONO/NO2 ratio rises around noon at peak solar radiation. I have two problems with this that the authors should address. First, any ratio with NO2 in the denominator will increase as NO2 is photolyzed at greater rates. Second – and this is the more serious concern – is that 3-D air quality models predict an increase in HONO/NO2 ratios in the late morning through noon, but they certainly aren't influenced by missing HONO sources (e.g. Figure 8 in http://dx.doi.org/10.1016/j.atmosenv.2015.04.048). While there may well be a significant unknown HONO source during the day, relying on HONO/NO2 ratios does not sufficiently make the case.

Response: Thanks for the comments.

For first problem, we agree that the greater rates of NO_2 can also increase the HONO/NO_2 ratio. If just considering of the photolysis of HONO and NO_2, both of which will convert to NO, the loss of HONO and the almost unchanged concentration of NO_x (NO_2+NO) will reduce the ratio HONO/NO_x. So we actually use the ratio HONO/NO_x to present the conversion of NO_x to HONO partly (please see Fig.1 and Fig. 2 in the revised manuscript).

For second problem, the increase of HONO/NO_x at daytime can result from: (1) the homogeneous reaction of NO and OH radical (R3); (2) the conversion of NO_2 to HONO (R4, R5); (3) other NO_x-independent sources. In the work of Couzo et al. (2015) (Figure 8 in http://dx.doi.org/10.1016/j.atmosenv.2015.04.048), when they only considered R3, the predicted daytime HONO/NO_2 can follow the time variation of the measured ratio but underestimate significantly, and after include the heterogeneous formation from NO_2 (R4, R5) and HNO_3 (R6), the simulated HONO/NO_2 was improved during daytime, but significantly contradicted with the observed value in the second half of the night. Until now, the heterogeneous reaction mechanisms (R4, R5, R6) are actually not clear yet, there are uncertainties involved with the parameterizations in various models, many simulation works still tend to underestimate HONO concentrations (Czader et al., 2012;Lee et al., 2016).

The missing source ($P_{unknown}$) defined in our study contains the heterogeneous processes mentioned above. We want to understand which mechanism might be more important based on our measurements. The source of HONO is divided into gas phase reaction (R3), combustion emission and unknown source $P_{unknown}$. So both the homogeneous formation and unknown source of HONO can increase the HONO/NO_x ratio at daytime, with a mean value of 0.71 ppb/h and 1.02 ppb/h, respectively. $P_{unknown}$ has found to correlated with NO_2*UVB, indicating the photo-induced heterogeneous conversion of NO_2 to HONO, but for now we do not have any solid evidence to identify which surface (ground surface and aerosol surface) are important in this potential mechanism.

Line 534:

$$P_{unknown} = J(HONO)[HONO] + k_{HONO+OH}[HONO][OH] + \frac{\nu_{HONO}}{H}[HONO] + \frac{\Delta HONO}{\Delta t} - k_{NO+OH}[NO][OH] - \frac{0.79\% \times \Delta NO_x}{\Delta t} \quad (10)$$

Line 261-264: If the HONO sources during daytime are consistent with those at night, the minimum HONO/NO_x ratios should occur at noon due to the intense photochemical loss of HONO. Therefore, there must be additional sources of HONO during daytime (e.g. R3).

Line 539-542: the average homogeneous reaction rate between NO and OH (P_{NO+OH}) is 0.71 ppb/h and P_{emis} just gives a tiny part of HONO at a rate of 0.02 ppb/h, meaning that most of HONO comes from an unknown source whose average rate ($P_{unknown}$) is 1.02 ppb/h, contributing about 58% of the production of HONO.

-Assuming the existence of a missing HONO source during the day, to what extent could it be explained by soil emissions?
Response: Thanks for the comment and suggestion.

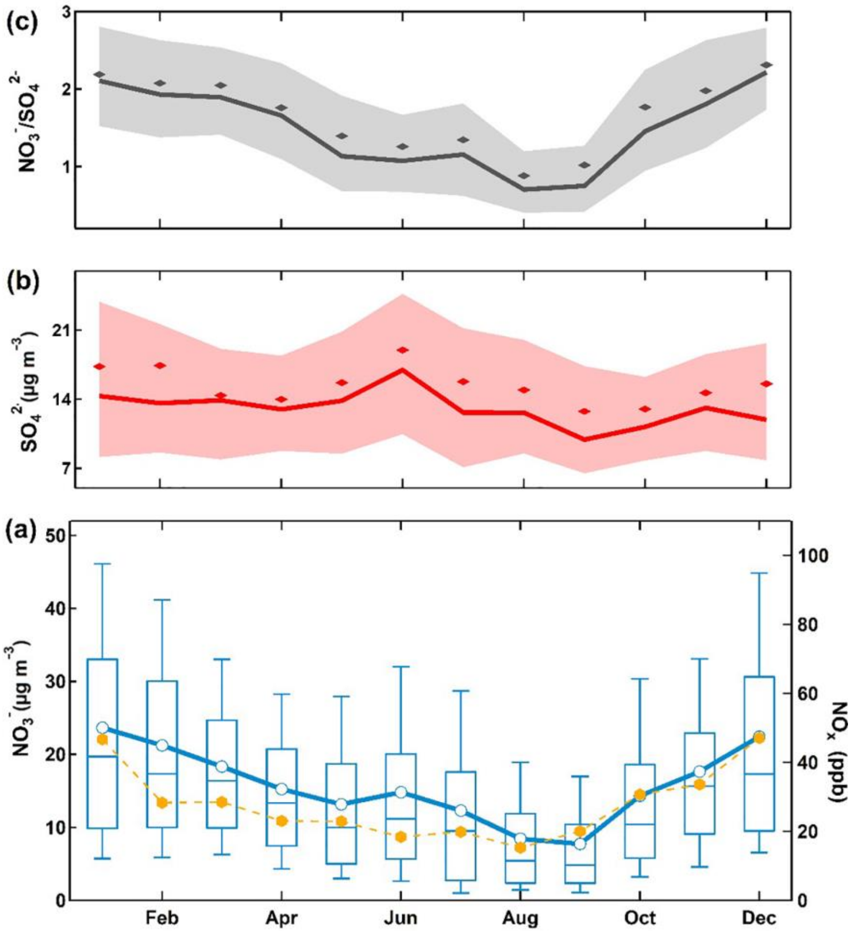
The averaged missing source calculated in our study is 1.02 ppb/h around noon (10:00-14:00 LT). So far, we cannot exclude the potential contribution from (photo-enhanced) heterogeneous reaction of NO_2, and the photolysis of adsorbed nitric acid (HNO_3) and particulate nitrate (NO_3^-). It's difficult to derive the rate or the amount of HONO emitted from soil emission, the main reason is that we were lack of direct observation. However, we are still trying to estimate the contribution of soil emissions to HONO through solving overdetermined equations at night, due to the relatively simple sources of HONO and without the influences of HONO photolysis, and the mixing effect of boundary layer (see part 4 in the revised manuscript for details). And, in average, 14.5% of nighttime HONO is found to be explained by soil emissions. The key to our calculation is the assumption that the mixing level of observed NH_3 can represent the intensity of soil emission of HONO. Although the processes of HONO and NH_3 emission from soil may not be completely synchronized, the seasonal patterns for each should be consistent.

Line 602-614: Although we do not directly measure HONO emissions from soil, the observed ammonia can represent its monthly average intensity, based on the following hypothesis: the *dominant source of NH_3 is from soil, especially from fertilizers ($NH_4+ \rightarrow NH_3$) for a good correlation between ammonia and temperature in the site (r=0.63, p=0.01), omitting the contributions of livestock to NH_3 since there is only a small poultry facility within 10 km of this site (Meng et al., 2011;Huang et al., 2012;Behera et al., 2013). Combustion sources (vehicles, industry, biomass burning) should contribute only a fraction of NH_3 seeing that NH_3 is not related to NOx or CO in our study. Moreover, the release of both HONO and NH_3 depend on the strength of microbial activities, fertilizing amount, and soil properties (e.g., temperature, acidity and water content of soil). Although the processes of HONO and NH_3 emission from soil may not be completely synchronized, the seasonal patterns for each should be consistent.*

-The authors state that mass concentration of PM2.5 is likely not the only factor affecting HONO formation on aerosol surfaces. This makes sense intuitively. Do the authors have speciated PM2.5 measurements during this time? How does the chemical composition of aerosols change throughout the year? Would these changes make the NO2 to HONO conversion more or less likely?

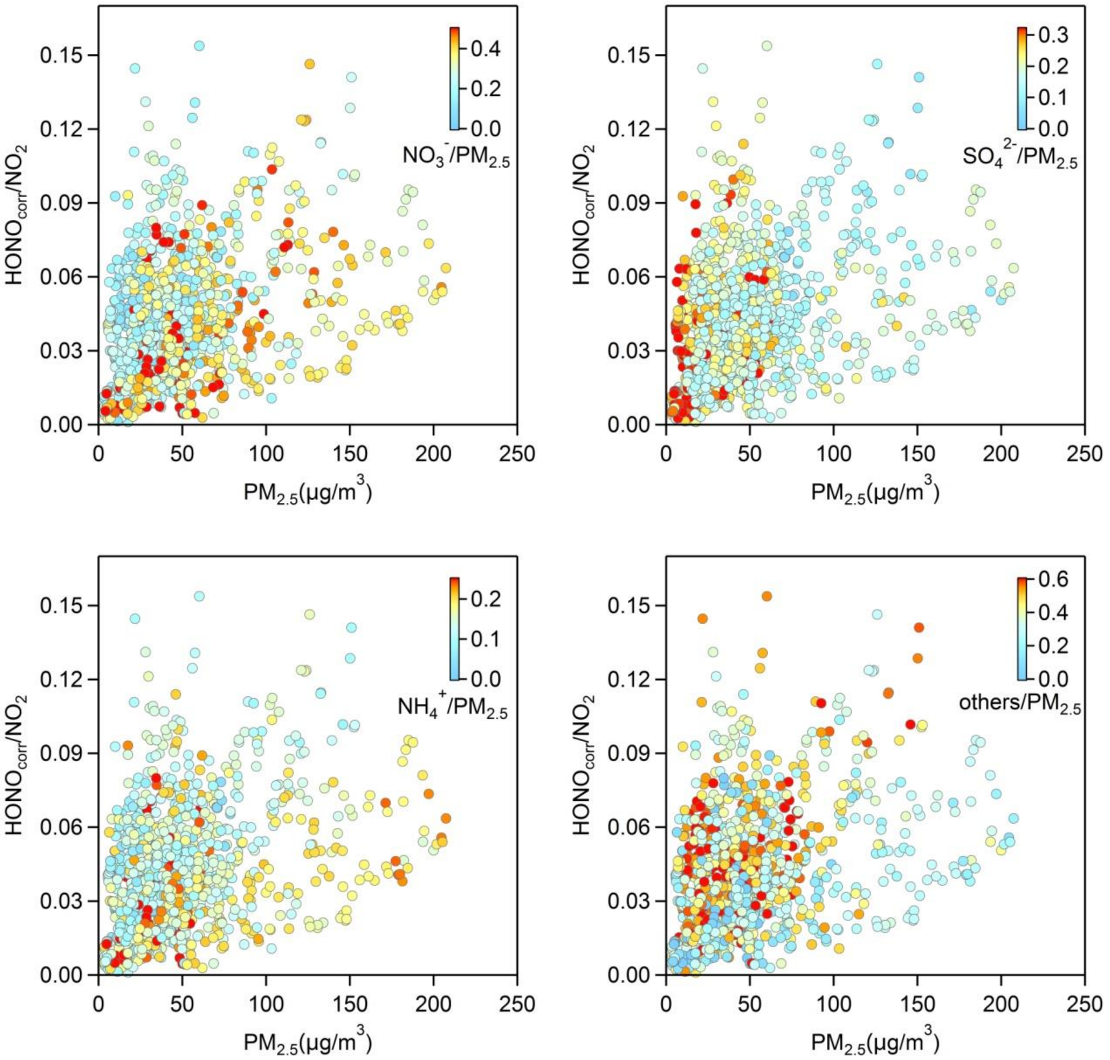
Response: Thanks for the comment and suggestion.

The seasonal variation of aerosol compositions has been reported in our previously work, showed in the following first figure: the particulate nitrate exhibits a maximum value in January and a minimum in August, and particulate sulfate shows a relatively weak seasonal cycle (Sun et al., 2018). An intuitive conclusion is that the proportion of nitrate will increase and the proportion of sulfate will decrease with the aerosol loading, from summer to winter.



Monthly averaged nitrate (blue), sulfate (red), NO_x (orange) mass concentrations and nitrate to sulfate molar-based ratio (grey) measured at the SORPES station during March 2014 to February 2016 (Sun et al., 2018).

The slope of $HONO_{corr}/NO_2$ and $PM_{2.5}$ varies over a relatively wide range, caused by some unknown factors that need to be explored. As the following figure shows, when the proportion of nitrate in aerosol is higher, the slope of $HONO_{corr}/NO_2$ and $PM_{2.5}$ tend to be lower slightly while the relationship shows differently for sulfate. The value of $(PM_{2.5}-NO_3^--SO_4^{2-}-NH_4^+)/PM_{2.5}$ can roughly represent the ratio of organic compounds in most situations, and it seems that the high ratio of organic aerosol occurs with the high slope of $HONO_{corr}/NO_2$ and $PM_{2.5}$. But simply relying on these cannot make too much sense, for example, the heat can make particulate nitrate volatilize into nitric acid gas and cause soil to emit more HONO, so we can see the highest $HONO_{corr}/NO_2$ ratio and the lowest proportion of nitrate to aerosol in summer. In future work, we're going to study the impact of aerosol components to the heterogeneous formation of HONO through laboratory experiments.



Scatter plot of $HONO_{corr}/NO_2$ and $PM_{2.5}$ in the time (3:00-6:00 LT) when $HONO_{corr}/NO_2$ reaches the pseudo steady state each night and are colored by the ratios of main aerosol compositions: (a) $NO_3^-/PM_{2.5}$, (b) $SO_4^{2-}/PM_{2.5}$, (c) $NH_4^+/PM_{2.5}$, (d) others/$PM_{2.5}$, i.e. $(PM_{2.5}-NO_3^--SO_4^{2-}-NH_4^+)/PM_{2.5}$.

Other minor comments:

-Check the in-text references to Figures and Tables. Some of the Figures are mis-referenced (e.g. referencing Fig 5 when, in fact, the figure being referenced is Fig 6). This happens quite often in the latter half of the manuscript.

Response: Thanks. We have re-edited the references to Figures and Tables in the revised manuscript.

- The last sentence in the second paragraph of section 3.3.2 is particularly confusing.

Response: Thanks. We have re-edited the language in the revised manuscript.

Line 393-398: Even at the lowest measured RH of 18%, the absolute moisture content in the atmosphere is still greater than 10^3 ppm in our study, which is quite abundant to react with NO_2, *but the $HONO_{corr}/NO_2$ ratio is quite small and remains unchanged when RH is below 45%, indicating that the NO_2 to HONO conversion efficiency should be determined by water covering the surfaces, rather than by the amount of water in the air.*

-To improve readability, try to have a native English speaker proofread the manuscript. Some of the phrases are oddly worded and obscure the authors' meaning.

Response: Thanks for the suggestion.

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