Itemized Response to Anonymous Referee #1's Comments

Ms. Ref. No.: acp-2019-916

Title: Characteristics, sources, and reactions of nitrous acid during winter at an urban site in the Central Plains Economic Region in China

Response to Anonymous Referee #1:

We have carefully addressed your comments on our manuscript and made necessary revisions of the previous manuscript. We sincerely thank you for valuable and constructive inputs. We believe that we have adequately addressed all of your comments and thus the current version has been greatly improved with those valuable comments and further English editing. The revised phrases/sentences/paragraphs are shown in the line number of the revised text.

The followings are our itemized replies to your comments.

General comments:

1. In general, the text can be followed. However, there are many awkward, unclear, redundant, unnecessary, ambiguous, and confusing phrases/statements (see the following tech/English comments). A professional expert must edit the text for clarity and for better flow before resubmission.

> Response: We sent the manuscript to a professional expert to enhance the readability of the manuscript. The revised portion has been highlighted with yellow color in the revised version (see the response of the following tech/English comments).

2. The rationale for your study is weak – need more elaboration. The fact that no study has been performed in Zhengzhou (L 129) does not justify the novelty of your study. You should have covered the setbacks of previous studies and state those tasks (including tables/figures) currently evaluated have not been properly addressed in previous studies. Unfortunately, I have found none. Anyway, pls emphasize the

uniqueness of your study.

Response: For your comment, we add the sentences in the revised text.

L 134-147: Many papers (Huang et al., 2017; Tong et al., 2016) took $PM_{2.5}$ as the main control factor of HONO, and studied the differences of HONO sources and characteristics between clean and polluted periods. Homogeneous reaction, direct emission, heterogeneous reaction, and daytime budget analysis were conducted during the period of worsening pollution (namely HD period in this paper). Total NO_X emissions in cities with different leading factors of emissions have been declining year by year due to Chinese government emission control measures, but some Chinese cities are still in high-NO_X areas (e.g. Beijing, Shanghai, Guangzhou, and Zhengzhou.) (Kim et al., 2015; Liu et al., 2017). Under high-NO_X conditions, some papers (Cui et al., 2018; Hou et al., 2016) suggested that heterogeneous reaction was the main source of HONO and did not conduct a quantitative analysis of homogeneous reaction, especially in winter. So, we explore relevant studies of homogeneous reactions. In addition, the source contributions of HONO at night varied with the degree of pollution level were not explained.

3. I have a hard time figuring out that the results from one single sampling site located on the rooftop of a building in Zhengzhou university (L 134) can represent the air quality in general and HONO in particular in the entire Zhengzhou city (180 million population, L 106). This is why there are so many monitoring stations (traffic, urban and background) within a given large city to reflect the air quality within the city. Pls modify your title as well those in the text.

Response: Thank you for your comment. As you say, one single sampling site cannot represent the air quality in general and HONO in the entire Zhengzhou city. We have revised the title, "Characteristics, sources, and reactions of nitrous acid during winter at an urban site in the Central Plains Economic Region in China".

4. Need to clearly cover HONO sources and sinks as well as homogeneous (NO+ •OH) and heterogenous ($NO_2 + H_2O$) in the text (introduction and discussion). For example, the role of ground surface at night for HONO deposit (sink) and the reemission (source) from HONO reservoirs (e.g., soil nitrite), etc. The other sink source, albeit insignificant, is that HONO may react with others to form new compounds, as in the case of reactions with amines in forming nitrosamines. How about transport of HONO? – what is the lifetime in atmosphere (hours under in-door conditions).

Response: OK. We added the sentence in the introduction in the revised text.

L 51-54: Therefore, the changes in the contribution of the homogeneous reaction, heterogeneous conversion, and direct emission during pollution can be observed by studying the formation mechanism of HONO.

In the discussion, we thought that the contribution of soot surface to HONO production is usually much lower than expected because the uptake efficiency of NO_2 decreases with the prolonged reaction time caused by surface deactivation. The aerosol surface is an important medium for the heterogeneous transformation from NO_2 to HONO (Liu et al., 2014). So, we added the sentences in the revised text.

L 364-368: the contribution of soot surface to HONO production is usually much lower than expected because the uptake efficiency of NO_2 decreases with the prolonged reaction time caused by surface deactivation. The aerosol surface is an important medium for the heterogeneous transformation from NO_2 to HONO (Liu et al., 2014).

We have clearly known that other HONO sources are not the main HONO sources and sinks: 1. HONO is formed by NO_2 through the photolysis of sooty surface and adsorbed nitric acid and nitrate at UV wavelengths (Kleffmann et al., 1999). 2. The homogeneous nucleation of NO_2, H_2O, and NH_3 is the HONO formation pathway (Zhang and Tao, 2010). 3. HONO can deposit and react with amines in forming nitrosamines (Li et al., 2012). So, we added the sentences in the revised text.

L 98-102: The unknown sources of HONO may include the NO_2 photolysis of sooty surface and adsorbed nitric acid and nitrate at UV wavelengths (Kleffmann et al., 1999). The homogeneous nucleation of NO_2, H_2O, and NH_3 is the HONO formation pathway (Zhang and Tao, 2010). In the meanwhile, HONO can deposit and react with amines in forming nitrosamines (Li et al., 2012) for sinking.

We knew that the lifetime of HONO was 10–20 min at daytime (Lu et al., 2018) and the estimated lifetime was about 3.3 h in the nighttime (Nie et al., 2015).

5. Need to discuss the uncertainty in your results due to variations of many parameters (e.g., rate constants and OH radical values, L 250-253). This leads to the following comments about the use of significant figures.

Response: Measured species and performance of the instruments are counted in **Table S1**. The rate constants are learned from the study (Atkinson et al., 2004). We don't know the uncertainty of rate constants.

Table S1.
Measured species and performance of the instruments.

Species	Measurement technique	Detection limit	Accuracy
$PM_{2.5}$	Tapered Element Oscillating Microbalance	1.5 μg m^{-3}	± 5%
HONO	Ion Chromatography	4 pptv	± 20%
CO	Absorbs Infrared Radiation	40 ppbv	± 5%
NO	Chemiluminescence	60 pptv	± 20%
NO_2	Chemiluminescence	300 pptv	± 20%
O_3	UV Photometry	0.5 ppbv	± 5%

The results came from instrument manufacturers.

Hence, we assumed ± 50% ·OH values to estimate the uncertainty of P_{OH+NO}^{net}. The ·OH values of 1.25×10^5 and 3.75×10^5 molecule cm^{-3} were calculated the P_{OH+NO}^{net} values of 0.16 and 0.49 ppbv h^{-1}.

L 295-297: We assumed ± 50% ·OH values to estimate the uncertainty of P^{net}_{OH+NO}. The ·OH values of 1.25×10^{5} and 3.75×10^{5} molecule cm^{-3} were calculated the P^{net}_{OH+NO} values of 0.16 and 0.49 ppbv h^{-1}.

6. Be careful about use of significant figures. Delete decimal points for RH (L 182, 185, 187, 194, Table 2, etc.), for NO2 level (L 197-198; also, you compare with standard of 80), for PM2.5 (Table 2), for level in μg m-3 (L 198, etc.), for ratio (L 34, 304, etc.).

Response: Thank you for the comment. We modified the problem in **Table 1** and revised the text. We have learned how to use significant figures.

Table 1.

Data statistics of HONO, $PM_{2.5}$, NO_2, NO, NO_X, HONO/NO_2, HONO/NO_X, O_3, CO, T, RH, and WS during the measurement period, mean value ± standard deviation.

Trace gases	CD			PD			SPD			Total days
	Day	Night	All	Day	Night	All	Day	Night	All	
$PM_{2.5}$ (μg m^{-3})	37 ± 15	41 ± 17	39 ± 16	80 ± 32	93 ± 46	87 ± 40	148 ± 29	147 ± 33	147 ± 31	91 ± 54
HONO (ppbv)	0.9 ± 0.7	1.4 ± 0.7	1.1 ± 0.7	1.9 ± 1.7	2.7 ± 1.3	2.3 ± 1.5	3.5 ± 2.7	4.0 ± 1.1	3.7 ± 2.1	2.5 ± 1.9
CO (ppmv)	1 ± 0.3	1 ± 0.3	1 ± 0.3	1± 0.4	1 ± 0.6	1 ± 0.5	2 ± 0.6	2 ± 0.4	2 ± 0.5	1 ± 0.6
NO (ppbv)	18.4 ± 39.3	15 ± 34.3	16.7 ± 36.8	20.3 ± 26.2	30.7 ± 33.6	25.5 ± 30.4	40.8 ± 50.8	64.3 ± 82.1	52.5 ± 68.9	31.8 ± 51.4
NO_2 (ppbv)	23 ± 13	26 ± 13	25 ± 13	29 ± 9	38 ± 10	33 ± 11	40 ± 11	43 ± 10	42 ± 11	33 ± 14
O_3 (ppbv)	21.4 ± 11.5	13.8 ± 10.0	17.6 ± 11.4	17.4 ± 11.9	8.9 ± 8.1	13.1 ± 10.9	15.6 ± 14.2	7.9 ± 7.1	11.8 ± 11.8	14.2 ± 11.7
HONO/NO_2 (%)	4.2 ± 3.6	5.3 ± 2.2	4.7 ± 3.1	6.8 ± 5.8	7.4 ± 3.9	7.1 ± 4.9	9.0 ± 7.7	9.8 ± 5.8	9.4 ± 6.8	7.6 ± 6.4
HONO/NO_X (%)	3.3 ± 2.7	6.0 ± 5.6	4.5 ± 4.5	4.4 ± 2.5	4.6 ± 1.7	4.5 ± 2.1	5.3 ± 3.4	5.8 ± 4.7	5.6 ± 4.1	4.9 ± 3.8
RH (%)	30 ± 21	36 ± 20	33 ± 21	44 ± 17	54 ± 18	49 ± 18	64 ± 18	73 ± 13	68 ± 16	50 ± 24
WS (m s^{-1})	0.8 ± 1.0	0.5 ± 0.7	0.7 ± 0.9	1.1 ± 1.4	0.6 ± 0.9	0.9 ± 1.2	0.4 ± 0.7	0.3 ± 0.6	0.4 ± 0.7	0.6 ± 0.9
T (°C)	4.3 ± 4.6	2.7 ± 3.6	3.5 ± 4.2	3.7 ± 3.3	2.6 ± 3.1	3.1 ± 3.2	4.6 ± 3.2	2.9 ± 2.1	3.8 ± 2.8	3.5 ± 3.5

Delete decimal points for RH (L182, 185, 187, 194, Table 2, etc.),

Response:

L 205: …with RH ranging from 5 to 79%...

L 207: …RH ranging from 17 to 86%...

L 210: …RH ranging from 30 to 96%...

L 217: …RH in CD, PD, and SPD periods was 33, 49, and 68%...

level in μg m-3 (L 198,etc.),

Response:

L 221: …values of NO_2 were 25, 33, and 42 ppbv (46, 63, and 78 μg m^{-3} lower than…

for ratio (L 34, 304, etc.).

Response:

L 34: …HONO ratio (less than 20%) was approximately 77%...

L 343: …$HONO_{emission}$/HONO ratio less than 20% was approximately 77%...

7. Be consistent with the format of unit. You use m s-1 (or μg m_{-3}), yet m/s and μg·m_{-3} are in Table 2 (delete centered dot). Use ppbv throughout the text, but ppbV in Tables 1 and 2 and Fig. 8 (some ppbv and one uses ppbV).

Response: Sorry for the careless. The problems have been revised. And we checked the full text to avoid the problems.

Table 1.

Data statistics of HONO, $PM_{2.5}$, NO_2, NO, NO_X, HONO/NO_2, HONO/NO_X, O_3, CO, T, RH, and WS during the measurement period, mean value ± standard deviation.

Trace gases	CD			PD			SPD			Total days
	Day	Night	All	Day	Night	All	Day	Night	All	
$PM_{2.5}$ (μg m^{-3})	37 ± 15	41 ± 17	39 ± 16	80 ± 32	93 ± 46	87 ± 40	148 ± 29	147 ± 33	147 ± 31	91 ± 54
HONO (ppbv)	0.9 ± 0.7	1.4 ± 0.7	1.1 ± 0.7	1.9 ± 1.7	2.7 ± 1.3	2.3 ± 1.5	3.5 ± 2.7	4.0 ± 1.1	3.7 ± 2.1	2.5 ± 1.9
CO (ppmv)	1 ± 0.3	1 ± 0.3	1 ± 0.3	1± 0.4	1 ± 0.6	1 ± 0.5	2 ± 0.6	2 ± 0.4	2 ± 0.5	1 ± 0.6
NO (ppbv)	18.4 ± 39.3	15 ± 34.3	16.7 ± 36.8	20.3 ± 26.2	30.7 ± 33.6	25.5 ± 30.4	40.8 ± 50.8	64.3 ± 82.1	52.5 ± 68.9	31.8 ± 51.4
NO_2 (ppbv)	23 ± 13	26 ± 13	25 ± 13	29 ± 9	38 ± 10	33 ± 11	40 ± 11	43 ± 10	42 ± 11	33 ± 14
O_3 (ppbv)	21.4 ± 11.5	13.8 ± 10.0	17.6 ± 11.4	17.4 ± 11.9	8.9 ± 8.1	13.1 ± 10.9	15.6 ± 14.2	7.9 ± 7.1	11.8 ± 11.8	14.2 ± 11.7
HONO/NO_2 (%)	4.2 ± 3.6	5.3 ± 2.2	4.7 ± 3.1	6.8 ± 5.8	7.4 ± 3.9	7.1 ± 4.9	9.0 ± 7.7	9.8 ± 5.8	9.4 ± 6.8	7.6 ± 6.4
HONO/NO_X (%)	3.3 ± 2.7	6.0 ± 5.6	4.5 ± 4.5	4.4 ± 2.5	4.6 ± 1.7	4.5 ± 2.1	5.3 ± 3.4	5.8 ± 4.7	5.6 ± 4.1	4.9 ± 3.8
RH (%)	30 ± 21	36 ± 20	33 ± 21	44 ± 17	54 ± 18	49 ± 18	64 ± 18	73 ± 13	68 ± 16	50 ± 24
WS (m s^{-1})	0.8 ± 1.0	0.5 ± 0.7	0.7 ± 0.9	1.1 ± 1.4	0.6 ± 0.9	0.9 ± 1.2	0.4 ± 0.7	0.3 ± 0.6	0.4 ± 0.7	0.6 ± 0.9
T (°C)	4.3 ± 4.6	2.7 ± 3.6	3.5 ± 4.2	3.7 ± 3.3	2.6 ± 3.1	3.1 ± 3.2	4.6 ± 3.2	2.9 ± 2.1	3.8 ± 2.8	3.5 ± 3.5

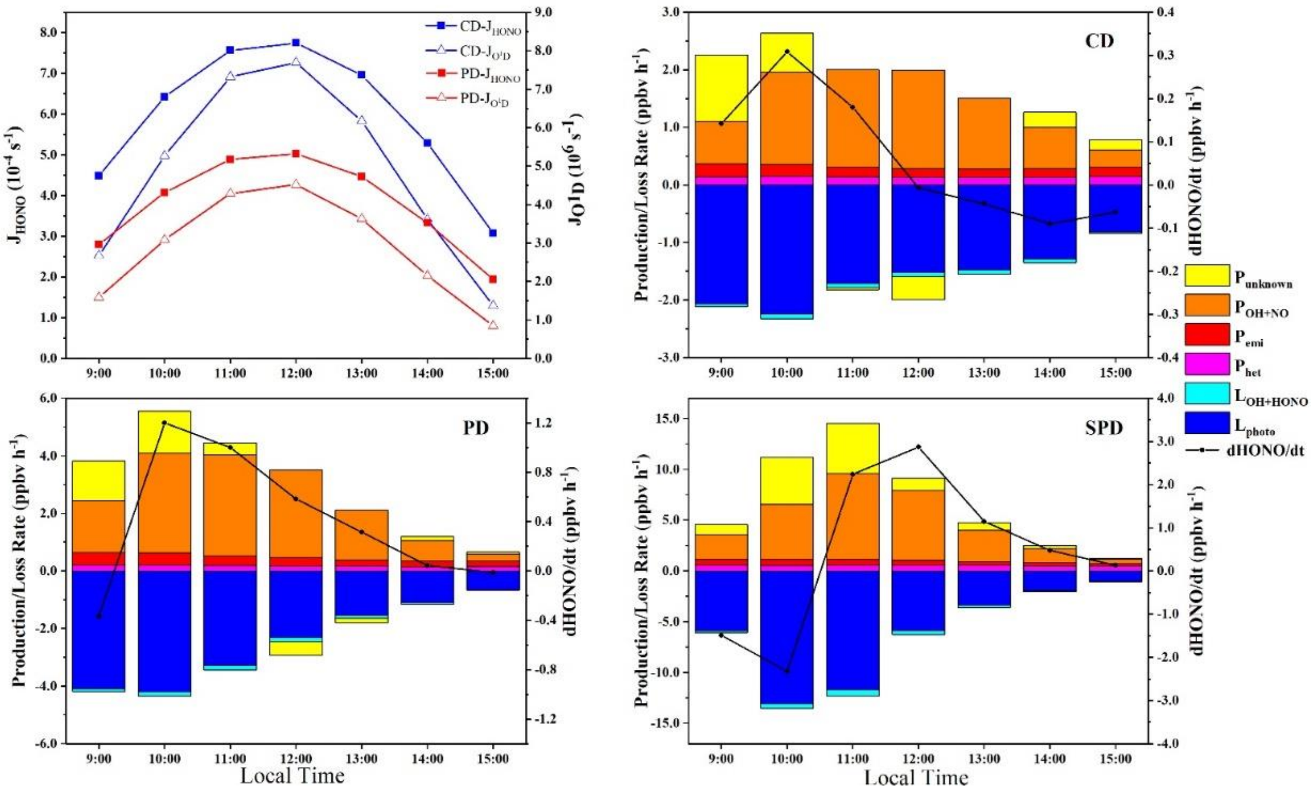

Fig. 9. The average profiles of J_{HONO} and J_{O^1D} concentrations during the daytime, and production and loss rate of the daytime HONO in CD, PD and SPD periods.

8. Data need show the variation; use Box plots or error bars in figures and add standard deviation in tables.

Response: OK. With too many error bars in figures, it will make the figures look unclear. So the error bars of **Fig. 4**, **Fig. 5**, and **Fig. 8** were placed separately in the tables of the supplement (**Table S2**, **Table S3**, and **Table S4**).

9. Need proper citations for equations and rate constants, e.g., in L 247, 250

Response: Sorry, it was our error of citations. Proper citations have been added. L 278: At T = 298 K and P = 101 kPa… respectively (Atkinson et al., 2004; Sander et al., 2003).

10. The comparison with others (Table 1) may not be useful and must be made with care since the studied year is different (some in 2012 – may not have adequate end-of-the pipe treatment), nature of sampling sites is different (some in urban, suburban and even rural sites) and atmospheric dynamics in these regions are far different.

Response: You're right. The comparison with others must be made with care since the studied year is different. Therefore, only the observation data of HONO in the last ten years were used for comparative analysis of HONO concentration changes in urban. We removed the HONO level (Jun.-Jul. 2005 in Germany) in the revised text (**Table 2**). We analyzed the effect of the site on HONO concentration in urban. So, we added sampling sites in **Table 2**.

Table 2.

Comparisons of the daytime and nighttime HONO level, $HONO/NO_2$, and $HONO/NO_X$ mean values in Zhengzhou and other sites around the world.

Date (Site)	Instrument	HONO (ppbv)			$HONO/NO_2$ (%)		$HONO/NO_X$ (%)		Reference
		Day	Night	N/D	Day	Night	Day	Night	
Oct.–Nov. 2014 (Beijing, urban)	LOPAP (long path absorption photometer)	0.9	1.8	2.0	2.6	4.6	1.7	2.2	Tong et al., 2015
Feb.–Mar. 2014 (Beijing, urban)	LOPAP	1.8	2.1	1.2	3.8 (Severe haze)	4.3	2.5	2.5	Hou et al., 2016
		0.5	0.9	1.8	7.8 (Clean)	3.0	5.1	2.4	
Jul. 2006 (Guangzhou, rural)	LOPAP	0.2	0.9	4.5	1.0	2.5	4.3	4.5	Li et al., 2012
Jul. 2014–Aug. 2015 (Xi'an, urban)	LOPAP	0.5	1.6	3.2	3.3	6.2			Huang et al., 2017
Aug. 2010–Jun. 2012 (Shanghai, urban)	Active DOAS	0.8	1.1	1.4	4.2	4.5			Wang et al., 2013
Jul. 2009 (Paris, urban)	wet chemical derivatization technique-HPLC/UV-VIS detection	0.1	0.2	2.0	3.3	2.5			Michoud et al., 2014
Jan. 2019	AIM	2.2	2.8	1.3	6.8	8.5	4.4	5.5	This study

Jul. 2006 (Guangzhou, urban): This paper is a relatively early study on the HONO concentration level in China. We thought this paper still had a certain level of high quality, so we put it in the table for comparison.

For avoiding this problem of atmospheric dynamics, we used observed values of HONO in Chinese cities mostly.

11. Why no discussion of OH production rate as a function of O_3 levels?? In other words, is any HONO information related to O_3 pollution level?? It is relevant since •OH radical generated from HONO is in turn used for O_3 production.

Response: Thank you for the comment. We hold the opinion that the discussion of OH production rate as a function of O_3 levels can be written as an article on account of complexity. At the same time, we found that the negative correlation between HONO and O_3 was lower in the entire period, and it was known from **Fig 4d** that the heavier the pollution level, the lower the O_3 concentration.

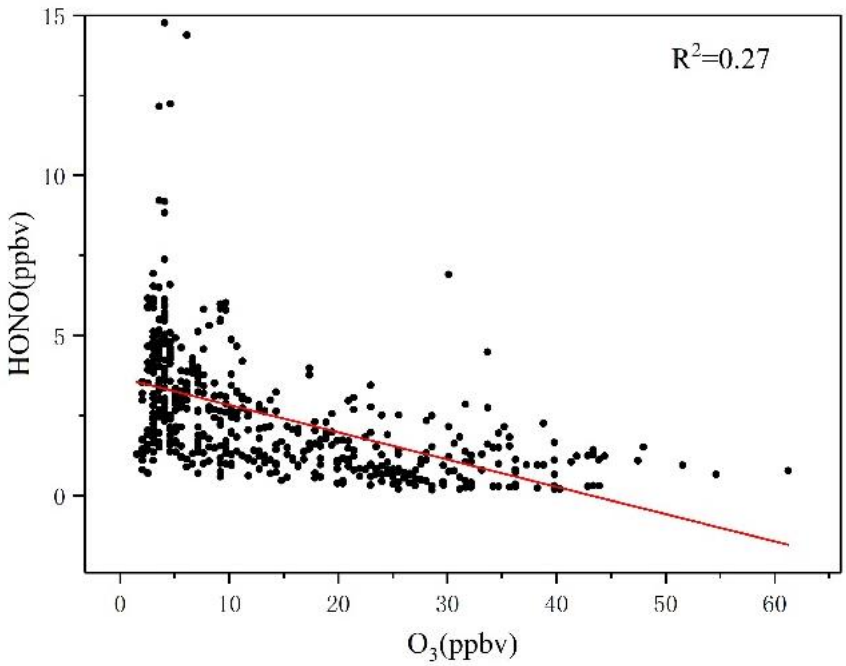

Figure The correlation between HONO and O_3.

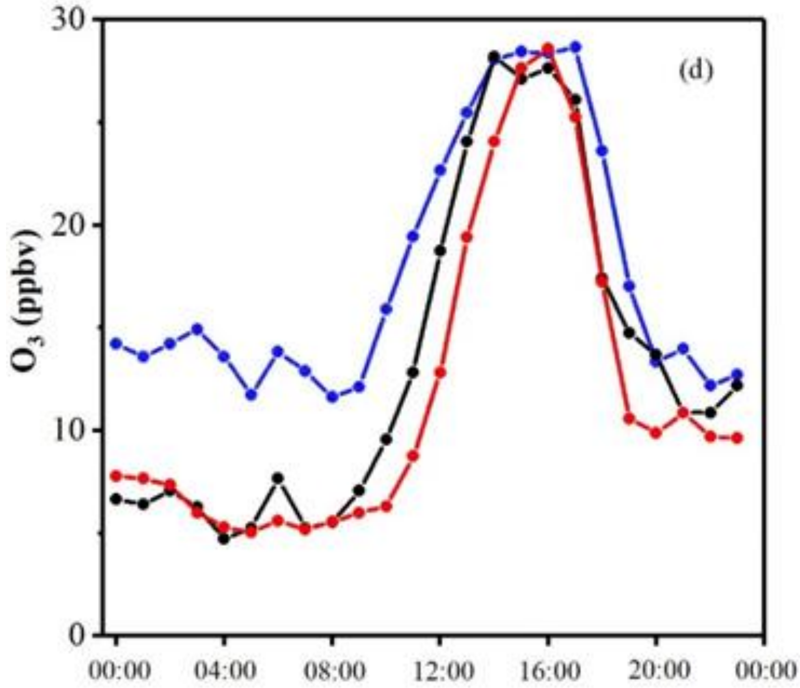

Fig. 4d. Diurnal variations of O_3. The blue points and lines represented the CD period; the black points and lines represented the PD period; the red points and lines represented the SPD period.

Specific comments:

1. “Title is misleading”

Response: OK. We have added “at an urban site” in the title. The title is “Characteristics, sources, and reactions of nitrous acid during winter at an urban site in the Central Plains Economic Region in China.”

2. L 15: All of a sudden, the phrase “three sources” pops up. Need to clearly state what they are.

Response: According to your comment, the sentence was modified in the revised text.

L 14-15: The contribution of the homogeneous reaction, heterogeneous conversion, and direct emission to HONO sources varied under different pollution levels.

3. Why not place centered dot symbol for OH radical? or •OH

Response: OK. We have changed ·OH for ·OH in the revised text.

L 44: …HONO to ·OH concentration…

L 47: …most important primary source of ·OH…

L 77: …react with the ·OH…

L 96: …the importance of the ·OH…

L 280: …concentration of ·OH…

L 282: …the average concentration of ·OH…

L 283: …the same ·OH…

L 471: …·OH and NO were formed…

L 495: …values of J_{HONO} and ·OH concentration…

4. Use a proper term in lieu of rate which refers to time

Response: Sorry for the careless. We have removed the word “rate”.

L 44: …the contribution of HONO to ·OH concentration can reach 25−50%…”

5. L 45: Why only these two? What about others including HONO itself and acetone?

Response: We have modified the sentence in the revised text.

L 43-46: …the contribution of HONO to ·OH concentration can reach 25%−50%, especially when the concentration of OH radicals produced by the photolysis of ozone, acetone, and formaldehyde…

6. L 49: what reaction? Should be more than one reaction.

Response: Sorry for my confusion. We have changed “reaction” for “the changes in the contribution of the homogeneous reaction, heterogeneous conversion, and direct emission” in the revised text.

L 51-54: …the changes in the contribution of the homogeneous reaction, heterogeneous conversion, and direct emission during pollution can be observed…

7. Citation:

Response: Sorry for my carelessness. All modifications have been made, and we have checked the full text.

a. Provide adequate spacing between citations, or Hou et al., 2016; Michoud et…. (need a spacing before the 2nd citation).

L 57: …(Elshorbany et al., 2012; Winer and Biermann, 1994)…

L 60: …(Duan et al., 2018; Min et al., 2016)…

L 61: …(Hirokawa et al., 2009; Roberts et al., 2010).

L 68: …(Hou et al., 2016; Michoud et al., 2014).

L 70: …(Acker et al., 2005; Grassian, 2001; Kurtenbach et al., 2001).

b. Make sure all cited references are listed and vice versa. For example, citations in L 55 (L 60) and Table 1 (Elshorbany et al., 2012) are not listed.

L 785: VandenBoer, T. C., Markovic…

L 606: Elshorbany, Y. F., Steil, B., Brühl, C…

c. Delete redundant citations. No need to cite twice in the beginning and at the end of the sentence. Pls change throughout the text (L 75, 82, 88, 93, 156, 157, 246, etc.) by delete the second citation.

L 72: Kurtenbach et al. (2001)…

L 77: Tong et al. (2015)…

L 90: Hao et al. (2006)…

L 96: Su et al. (2008a)…

L 105: Spataro et al. (2013)…

L 108: Tong et al. (2015)…

d. L 95; et al. (2013); delete the extra comma.

L 105: Spataro et al. (2013)…

e. Avid excessive self-citations (one is enough, e.g., L 115, 116, etc.)

f. For the same last name, use the format of Jiang et al., 2018c, 2018e; Liu…. (L 115). Pls change throughout the text.

L 123-127: As the core city of CPER, Zhengzhou characterized by severe PM (particulate matters) pollution (Jiang et al., 2018b), is selected in the study. In recent years, comprehensive PM research has been conducted on the chemical characteristics of PM in Zhengzhou. (Li et al., 2019), source apportionment (Liu et al., 2019), health risks (Jiang et al., 2019), and emission source profiles (Jiang et al., 2018a).

g. L 140: why cite this? Need year for this reference.

L 163-164: …(7:00–18:00 local time)…

h. L 181: why cite this one? Delete

L 204: …Air Quality Standards (CNAAQS) (75 $\mu g\, m^{-3}$)…

i. L 313: Acker et al. (2005) reported...

L 352: …Acker et al. (2005)…

8. L 60: what is 1:1??

Response: Sorry for my confusion. Through the comparison of measurement results, the correlation between SC-AP deployed onsite and AIM tended to 1. For avoiding the confusion, we have removed the words "; the results exhibited a consistency of nearly 1:1" in the revised text.

L 64-66: Compared with HONO measured by SC-AP deployed onsite, HONO measured by AIM has a small error and is within the acceptable analytical uncertainty (VandenBoer et al., 2014).

9. L 79: There is no connection between these two sentences. Need a sentence(such as revised R3 reaction) leading to the following sentence.

Response: OK. We have added the sentence between these two sentences in the revised text.

L 82-87: Such calculations have been applied in studies on homogeneous reactions and daytime budgets (Hou et al., 2016; Huang et al., 2017). These are studies of homogeneous reactions, and some researchers have begun to explore the mechanism of NO_2 heterogeneous reactions. Finlayson-Pitts et al. (2003) studied the mechanism of chemical adsorption of NO_2 and H ions on the adsorbed surface…

10. L 123: These two measurements ($PM_{2.5}$ and HONO) cannot clarify the sources, sinks and reactions. Pls reword.

Response: Sorry, we have modified the sentence in the revised text.

L 133-134: The levels of $PM_{2.5}$ were divided into three periods to analyze the HONO sources, sinks, and reactions in different periods.

11. L 173: what is uncertainty of AIM? How about MDL for other gases??

Response: Thank you for your comment. Measurement technique, detection limit, and accuracy of measured species are shown in Table S1 in the supplement.

Table S1.

Measured species and performance of the instruments.

Species	Measurement technique	Detection limit	Accuracy
$PM_{2.5}$	Tapered Element Oscillating Microbalance	1.5 μg m^{-3}	± 5%
HONO	Ion Chromatography	4 pptv	± 20%
CO	Absorbs Infrared Radiation	40 ppbv	± 5%
NO	Chemiluminescence	60 pptv	± 20%
NO_2	Chemiluminescence	300 pptv	± 20%
O_3	UV Photometry	0.5 ppbv	± 5%

The results came from instrument manufacturers.

12. L 190: Table 1 must come before Table 2. Rearrange the table number.

Response: OK. We have rearranged the table number.

L 213: **Table 1** lists the data statistics…

L 265: …listed in **Table 2.**

13. Interesting. You cover all these parameters (L 199-204) shown in Fig. 2, yet Fig. 2 is mentioned several sentences later (L205). The same illogical sequence is found in L 403 which mentions $P_{unknown}$ and P_{emi}, but the equation for these is shown much later (L 436). Also need to cover these rates for estimating daytime HONO budget.

Response: Sorry for my confusion. The illogical sequence had been changed. We have modified the sentences in the revised text.

L 223-226: **Fig. 2** shows the concentration changes in HONO…The variations of the average HONO, $PM_{2.5}$, NO_2…

L 456-465: …d HONO / d t = sources − sinks

$$= (P_{unknown} + P_{OH+NO} + P_{emi} + P_{het}) - (L_{OH+HONO} + L_{photo}) \quad (4),$$

$$P_{OH+NO} = k_{OH+NO} [OH] [NO] \quad (5),$$

$L_{OH+HONO} = k_{OH+HONO}$ [OH] [HONO] (6).

The d HONO / d t calculated from…

14. L 240: why??

Response: Sorry for my carelessness. Although observations of HONO levels in Zhengzhou were different from other cities because of periods, seasons, and meteorological conditions, a higher concentration of HONO was found in Zhengzhou. This situation attracted our attention. We thought the environmental impact of the increase in pollutant emissions and the number of vehicles exceeds the efforts of Zhengzhou to protect the atmospheric environment. Total NO_X emissions in cities have been declining year by year due to Chinese government emission control measures, but some Chinese cities are still in high-NO_X areas (e.g. Beijing, Shanghai, Guangzhou, and Zhengzhou.) (Kim et al., 2015; Liu et al., 2017). Therefore, we have added the sentence in the full text.

L 267-269: The reason for this phenomenon is that Zhengzhou is a high-NO_X area which provides HONO with abundant precursors (NO_2 and NO) in winter (Kim et al., 2015).

15. L 252: why in reference to Beijing?

Response: We revisit and determine the OH concentration. The OH concentration previously used is 1×10^6 molecule cm^{-3} is obtained by the simulation (Lelieveld et al., 2016). So, we have modified the sentences in the revised text.

L 280-285: [OH] is the concentration of ·OH that was not measured during the campaign. Therefore, Tan et al. (2018) found that by the field measurement, the average concentration of ·OH in Beijing at nighttime was about 2.5×10^5 molecule cm^{-3}. Moreover, the same ·OH concentration was also used to calculate the homogeneous reaction of HONO in the recent researches of Beijing (Zhang et al., 2019), Shanghai (Cui et al., 2018), and Xi'an (Huang et al., 2017).

Finally, we chose the average concentration of ·OH in Beijing was about 2.5×10^5 molecule cm^{-3} as the nighttime ·OH concentration in Zhengzhou.

16. L 253: Can you calculate OH radical concentration from those discussed later in L 418?

Response: OK. The concentration of ·OH during the daytime was calculated by the TUV model. But there is no sunlight at night, so it cannot be counted.

17. L 287: You are talking about night and mentioned no pollution source near the site. Why all these calculations related to traffic?? Unclear.

Response: Sorry for your comment. The sentence is not clear enough. So, we have changed "because no pollution source was near the measurement site" for "because the site is close to the western Fourth-Ring Expressway of Zhengzhou City and about Lian Huo Expressway to the north." in the revised text.

L 324-326: In the current study, directly emitted HONO could have been generated by vehicle exhaust and biomass combustion because the site is close to the western Fourth-Ring Expressway of Zhengzhou City and about Lian Huo Expressway to the north.

18. Pls explain the contradictory statements: important pathway (L 321) and unimportant pathway (L 20) for heterogeneous reaction for HONO formation.

Response: Sorry for my carelessness. With respect to direct emissions, heterogeneous reactions may be a more important pathway for HONO production.

The HONO/NO_2 ratio calculated in this work is much larger than that calculated for direct emission (< 1%) (Kurtenbach et al., 2001), suggesting that heterogeneous reactions may be a more important pathway for HONO production than direct emissions.

However, the average conversions of NO_2 (C_{HONO}) in CD, PD, and SPD periods were 0.72×10^{-2}, 0.64×10^{-2}, and 1.54×10^{-2} h^{-1}, respectively, indicating that the heterogeneous conversion of NO_2 was unimportant than the homogeneous reaction. So, in order to prevent confusion, we have modified the sentence in the revised text.

L 17-20: The average conversions of NO_2 (C_{HONO}) in CD, PD, and SPD periods were 0.72×10^{-2}, 0.64×10^{-2}, and 1.54×10^{-2} h^{-1}, respectively, indicating that the heterogeneous conversion of NO_2 was unimportant than the homogeneous reaction.

19. Use the stack format for equations in a separate line (L 389); use the solidus format for those within the line (L 402).

Response: Thank you. The equations had been changed.

L 438: $C_{HONO} = \frac{([HONO_{correct}]_{t2} - [HONO_{correct}]_{t1}}{(t2 - t1)\ [NO_2]}$

L 456: d HONO / d t = sources − sinks

20. L 431: Why the same values for both PD and SPD? That means you treat PD and SPD the same.

Response: OK. These are the averages per hour of J_{HONO} and J_{O^1D} for PD and SPD. We treated PD and SPD the same. The reason is that the main input parameters of TUV cannot be obtained directly, so we quoted the input parameters in the literature. However, the input parameters of PD and SPD are not distinguished in the papers. We wanted to study that under the same output conditions from the TUV model, the impact of different pollution levels changed on the daytime budget. We have added the sentence in the revised text.

L 491-493: We wanted to study that under the same output conditions from the TUV model in the PD and SPD periods, the impact of different pollution levels changed on the daytime budget.

21. Fig. 1: How could one tell the wind direction?? There is no shade area of black and red color (caption says so). Change in Fig. 2 too.

Response: Sorry. We have removed the word, "WD" in **Fig. 1** in the revised text. The colors of the shaded area were redefined in **Fig. 1** and **Fig. 2**. The shaded areas: white for the CD period; gray for the PD period; red for the SPD period.

22. Fig. 6d and RH effect: Was the phenomenon also observed by others?? Who is to say that 77% is the inflection point? Just say "reach a certain high level,HONO…."

Response: Thank you for your comment. The phenomenon also observed by other studies (Cui et al., 2018; Huang et al., 2017; Tong et al., 2015). "52% and 77%" was removed in the revised text.

L 400-402: When RH was increased, the $HONO/NO_2$ ratio began to increase rapidly with RH. The $HONO/NO_2$ ratio decreased when RH reached a certain high level.

L 409-410: However, the surface reached saturation when RH reached a certain high level.

23. References:

Response: Sorry for my carelessness. The problems had been changed in the revised text.

- Be consistent! Use lowercases for journals (e.g., L 502, 515, etc.)
- Need periods after all journal abbreviations (e.g., L 502, 515, etc.)

L 565: …Atmos. Res., 74, 507-524…

L 569: …J. Geophys. Res.,…

- Adequate subscript/superscript (e.g., L 535)

L 582: …gas phase reactions of O_x, HO_x, NO_x, and SO_x…

- Use correct journal, or Phy. Chem. Chem Phy., (L 536)

L 583: …Atmos. Chem. Phys.,...

- Use lowercases for articles (e.g., L 505)

L 563: …Concentrations of nitrous acid, nitric acid, nitrite and nitrate in the gas and aerosol…

- L 546 ref is not cited.

The reference has been removed in the revised text.

Tech/English comments:

Response: Thank you for your carefulness. The problems have been modified and modified issues have been marked in yellow throughout the revised text. The parts that need explanation have been listed.

1. L 13, 178, 179: Use polluted (not pollution)

L 12: …polluted days (PD), and severely polluted days…

L 200-201: …clean days [CD], polluted days [PD], and severely polluted…

2. L 33: Use was (not should be)

L 33: …OH + HONO, was at least 0.22 ppbv h^{-1}…

3. Be consistent about the use of verb tense; some use past tense in the first part of the sentence with present tense used in the latter part.

L 34-35: …(less than 20%) was approximately 77%, which suggested…

4. L 32, 279: Delete the redundant abatement (first one)

L 32-33: …hourly level of HONO abatement pathways…

5. L 44: delete rate

L 43-44: …the contribution of HONO to ·OH…

6. L 54: UV/Vis

L 58-59: …stripping coil-UV/Vis…

7. L 56: result comparison

L 62: A result comparison of different instruments…

8. L 67: have discovered

L 72: …have discovered that motor vehicles…

9. L 68-69: How does ratio "account for"? Use "is" 0.1-0.8%

L 74: …(aside from NO_X and other pollutants) is 0.1–0.8%.

10. Reword awkward L 81, word "radiation" (L 93), confusing (L 108-110).

L 85-87: Finlayson-Pitts et al. (2003) studied the mechanism of chemical adsorption of NO_2 and H ions on the adsorbed surface was revealed by using isotope-labeled water.

L 95-98: Su et al. (2008) revealed the importance of the ·OH from HONO during daytime (9:00–15:00 local time) and found that many unknown sources which are closely related to the solar radiation leading to HONO formation.

L 119-123: The file described the different factors which affect atmospheric pollution, including the level of economic development, energy structure, industrial structure and geographical location (solar radiation) with the Yangtze River Delta, Pearl River Delta, and Jing-Jin-Ji region.

11. L 99: Use pathway of…: mechanism is the same, but pathway is different.

L 109: …the pathway of HONO formation mechanism, namely…

12. L 107: "Food Production and Modern Agriculture" specified by (not published)

This website can not be found because of the update. We have changed "http://www.ndrc.gov.cn/zcfb/zcfbtz/201212/P020121203614181974825.pdf" for "http://www.gov.cn/zhengce/content/2011-10/07/content_8208.htm", in the revised text (L 119).

13. L 112: delete "is" and insert "is" ahead of selected

L 124: …is selected in the study.

14. L 114: Not Zhengzhou chemical characteristics. Should have written as: chemical characteristics of PM in Zhengzhou.

L 125-126: …on the chemical characteristics of PM in Zhengzhou…

15. L 121: $PM_{2.5}$ is not chemical

We have changed "chemical" for "factors" in the revised text (L 124).

L 131: …between HONO and other factors, such as $PM_{2.5}$…

16. L 124: How systematic?

We have removed "This investigation of $PM_{2.5}$ and HONO is expected to clarify the sources, sinks, and reactions in fine PM pollution and the importance of systematic research." We have added the sentences in the revised text.

L 133-147: The levels of $PM_{2.5}$ were divided into three periods to analyze the HONO sources, sinks, and reactions in different periods. Many papers (Huang et al., 2017; Tong et al., 2016) took $PM_{2.5}$ as the main control factor of HONO, and studied the differences of HONO sources and characteristics between clean and polluted periods. No homogeneous reaction, direct emission, heterogeneous reaction, and daytime budget analysis were conducted during the period of worsening pollution (namely HD period in this paper). Total NO_X emissions in cities with different leading factors of emissions have been declining year by year due to Chinese government emission control measures, but some Chinese cities are still in high-NO_X areas (e.g. Beijing, Shanghai, Guangzhou and Zhengzhou.) (Kim et al., 2015; Liu et al., 2017). Under high-NO_X conditions, some papers (Cui et al., 2018; Hou et al., 2016) suggested that heterogeneous reaction was the main source of HONO and did not conduct a quantitative analysis of homogeneous reaction, especially in winter. So, we explore relevant studies of homogeneous reactions. In addition, the source contributions of HONO at night varied with the degree of pollution level were not explained.

17. L 137: western Fourth-Ring Expressway

L 161: …from the western Fourth-Ring Expressway of Zhengzhou City…

18. L 151: Use chemicals (in lieu of substances)

L 173: The chemicals that could be oxidized…

19. L 153: O_2 and N_2 (not O and N)

L 174: …several gases (e.g., O_2 and N_2)…

20. L 160-161: Clearly state which instrument is for which compound, e.g., 48i for CO measurement.

We have modified the sentence in the revised text.

L 181-184: A temporal resolution of the model analyzer (TE [used for measuring O_3], 48i [used for measuring CO], 42i [used for measuring NO, NO_X, and NO_2], and TEOM 1405 $PM_{2.5}$ monitor [used for measuring $PM_{2.5}$], Thermo Electron, USA) is 1 h.

21. L 167-168: The manual uses the term “should be”. But in your statement, you should use “was changed” ….”was calibrated”.

L 193: The standard curve should be…

L 190-191: …were changed before the observation process, and the sampling flow was calibrated…

There are no serial **Numbers 22 and 23**. In order to prevent the confusion, the serial number used is consistent with the serial number in your comment.

24. L 171: A space after “≥” sign

L 194: …the correlation coefficient (≥ 0.999)…

25. L 181: specify that it is daily average

L 203: …the daily average of second grade…

26. Delete the first unit (L 182, 185, 188, 206, 301, 317, 360, etc.), first two units (L 301, 449, etc.)

L 205: …0 to 4.2 m s^{-1}…

L 208: …0 to 4.6 m s^{-1}…

L 210: …0 to 3.5 m s^{-1}…

L 230: …0.2 to 14.8 ppbv…

L 340: …2–52%, 6–34%, and 2–41%…

L 356: …1.3 and 59.0%…

L 401: …when RH reached a certain high level.

L 511: …15, 14, and 28%...

27. L 189: .. north with high WS … Also, how high is high? > 3 m/s, or > 4 m/s? Be specific!

L 212: …the maximum WS reached 4 m/s…

28. L 190: effect of pollutant removal

L 213: …the effect of pollutant removal…

29. L 202: mean values of what? of all pollutant concentrations?

L 226: The mean values of all pollutant concentrations except O_3…

30. L 209: Is your sampling site in urban area? You mentioned on the university campus. If so, the comparison is not valid.

OK. We have added the sampling site in Table 2. The university campus is urban in Zhengzhou. So, we compared the difference of the daytime and nighttime HONO level, HONO/NO_2, and HONO/NO_X mean values in urban in other cities.

Table 2.

Comparisons of the daytime and nighttime HONO level, $HONO/NO_2$, and $HONO/NO_X$ mean values in Zhengzhou and other sites around the world.

Date (Site)	Instrument	HONO (ppbv)			$HONO/NO_2$ (%)		$HONO/NO_X$ (%)		Reference
		Day	Night	N/D	Day	Night	Day	Night	
Oct.–Nov. 2014 (Beijing, urban)	LOPAP (long path absorption photometer)	0.9	1.8	2.0	2.6	4.6	1.7	2.2	Tong et al., 2015
Feb.–Mar. 2014 (Beijing, urban)	LOPAP	1.8	2.1	1.2	3.8 (Severe haze)	4.3	2.5	2.5	Hou et al., 2016
		0.5	0.9	1.8	7.8 (Clean)	3.0	5.1	2.4	
Jul. 2006 (Guangzhou, rural)	LOPAP	0.2	0.9	4.5	1.0	2.5	4.3	4.5	Li et al., 2012
Jul. 2014–Aug. 2015 (Xi'an, urban)	LOPAP	0.5	1.6	3.2	3.3	6.2			Huang et al., 2017
Aug. 2010–Jun. 2012 (Shanghai, urban)	Active DOAS	0.8	1.1	1.4	4.2	4.5			Wang et al., 2013
Jul. 2009 (Paris, urban)	wet chemical derivatization technique-HPLC/UV-VIS detection	0.1	0.2	2.0	3.3	2.5			Michoud et al., 2014
Jan. 2019	AIM	2.2	2.8	1.3	6.8	8.5	4.4	5.5	This study

31. L 214: No logic about sunrise – previous max HONO values (8-10 am) are way past sunrise. Should have written after 10 am…..

L 240: After 10:00 LT, the HONO concentration…

32. L 215: reword the incomplete phrase

We have reworded the sentence in the revised text.

L 240-242: After 10:00 LT, the HONO concentration decreased because of the increased solubility and rapid photolysis, remaining at a low level before sunset (14:00–16:00 LT).

33. L 219: delete again

L 243-244: After sunset, the concentrations of NO and NO_2 began to increase and remained at a higher level than the daytime.

34. L 220: and the concentration remained the same (Is it true??) until sunrise

It was not the same. The concentrations of NO and NO_2 began to increase and remained at a higher level than the daytime. We have modified the sentence in the revised text.

L 243-244: After sunset, the concentrations of NO and NO_2 began to increase and remained at a higher level than the daytime.

35. L 222: you meant NO_2 diffusion?

Sorry for my confusion. The concentrations of NO and NO_2 decreased after the peak values. On the one hand, NO and NO_2 can be involved in the reactions. On the other hand, NO and NO_2 diffused because of the boundary layer height increased in the daytime.

36. L 228: How is atmospheric migration? Should be "migration of atmospheric airmass".

L 253: …less affected by the migration of atmospheric airmass…

37. L 248: should be Eq. (1); Eq. (2) is in L 296.

L 277: …P_{OH+NO}^{net} = k_{OH+NO} [OH][NO] – $k_{OH+HONO}$ [OH][HONO] (1).

38. L 249, 337, 338, 349, 389, etc.: A space b/a the = sign

L 278: At T = 298 K and P = 101 kPa…

L 377-378: …$PM_{2.5}$ (R^2 = 0.23) was weaker than that between HONO and $PM_{2.5}$ (R^2 = 0.55)…

L 390-392: …CO was relatively moderate (R^2 = 0.43)…

L 438: …$C_{HONO} = \frac{([HONO_{correct}]_{t2} - [HONO_{correct}]_{t1}}{(t2 - t1)\,[NO_2]}$ (3)…

39. L 254: Adequate subscript in k_{OH+bar}

L 289: … the reaction rates of k_{OH+NO}…

40. L 274: reword

L 311-312: With the increase in pollution level, the HONO accumulation period at nighttime increased.

41. L 279: Use rate (not level)

L 316: …the hourly rate of HONO…

42. L 280: delete the extra spacing b/a the "/" sign

L 317-318: …3.36 – 1.59 ppbv)/8 h).

43. L 307: on the campus

L 346: …from the highway on the campus…

44. L 324: medium of what??

Sorry for my confusion. We have modified the sentence in the revised text.

L 361-364: With regard to the heterogeneous conversion of NO_2, several studies (An et al., 2012; Shen and Zhang, 2013) have reported that the surface of soot particles is the medium of NO_2 conversion.

45. L 349-350: wording? cannot see “indicating ...of what most cases”? Why?

Sorry for my confusion. The correlation coefficient between HONO and CO was relatively moderate (R^2 = 0.43), indicating that HONO and CO could come from the same source of emissions. Generally speaking, CO and NO are mainly related to combustion processes such as vehicle emissions, fossil fuel and biomass combustion (Tong et al., 2016). We have modified the sentence in the revised text.

L 390-396: The correlation coefficient between HONO and CO was relatively moderate (R^2 = 0.43), indicating that HONO and CO could come from the same source of emissions. Generally speaking, CO and NO are mainly related to combustion processes such as vehicle emissions, fossil fuel and biomass combustion (Tong et al., 2016). Thus, fossil fuel and biomass combustion may contribute to HONO production, but they can not be measured directly.

46. L 361: sedimentation of what? Do you mean deposit?

Right. We have modified the sentence in the revised text.

L 406-408: When RH ranged at the middle level, the heterogeneous conversion of NO_2 to HONO was more significant than that of deposition.

47. L 367: Delete “study of the”

L 414: The correlation between $HONO_{correct}$ and NO_2…

48. L 374: decreases after…

L 421: …heterogeneous reaction (R4), and NO_2 decreased after midnight.

49. L 383: in the (not then)

L 432: …calculated in the current study…

50. L 396: How is rate improvement? Use increase

OK.

L 445: The increase in the conversion rate demonstrates…

51. L 402: The expression of …. represents

L 454: The expression of d HONO / d t represents…

52. L 411: delete an

L 471: …·OH and NO were formed as R1.

53. Table 1: A space before (%), or HONO/NOX (%); before year, or Jun. 2012, Also add SD

Table 2.
Comparisons of the daytime and nighttime HONO level, $HONO/NO_2$, and $HONO/NO_X$ mean values in Zhengzhou and other sites around the world.

Date (Site)	Instrument	HONO (ppbv)			$HONO/NO_2$ (%)		$HONO/NO_X$ (%)		Reference
		Day	Night	N/D	Day	Night	Day	Night	
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Feb.–Mar. 2014 (Beijing, urban)	LOPAP	1.8	2.1	1.2	3.8 (Severe haze)	4.3	2.5	2.5	Hou et al., 2016
		0.5	0.9	1.8	7.8 (Clean)	3.0	5.1	2.4	
Jul. 2006 (Guangzhou, rural)	LOPAP	0.2	0.9	4.5	1.0	2.5	4.3	4.5	Li et al., 2012
Jul. 2014–Aug. 2015 (Xi'an, urban)	LOPAP	0.5	1.6	3.2	3.3	6.2			Huang et al., 2017
Aug. 2010–Jun. 2012 (Shanghai, urban)	Active DOAS	0.8	1.1	1.4	4.2	4.5			Wang et al., 2013
Jul. 2009 (Paris, urban)	wet chemical derivatization technique-HPLC/UV-VIS detection	0.1	0.2	2.0	3.3	2.5			Michoud et al., 2014
Jan. 2019	AIM	2.2	2.8	1.3	6.8	8.5	4.4	5.5	This study

The values of SD were shown in the references.

54. Fig. 3: Need errors bars. Use the present tense, “represent”. Show one example in Box plots so one has an idea about the magnitude of variation.

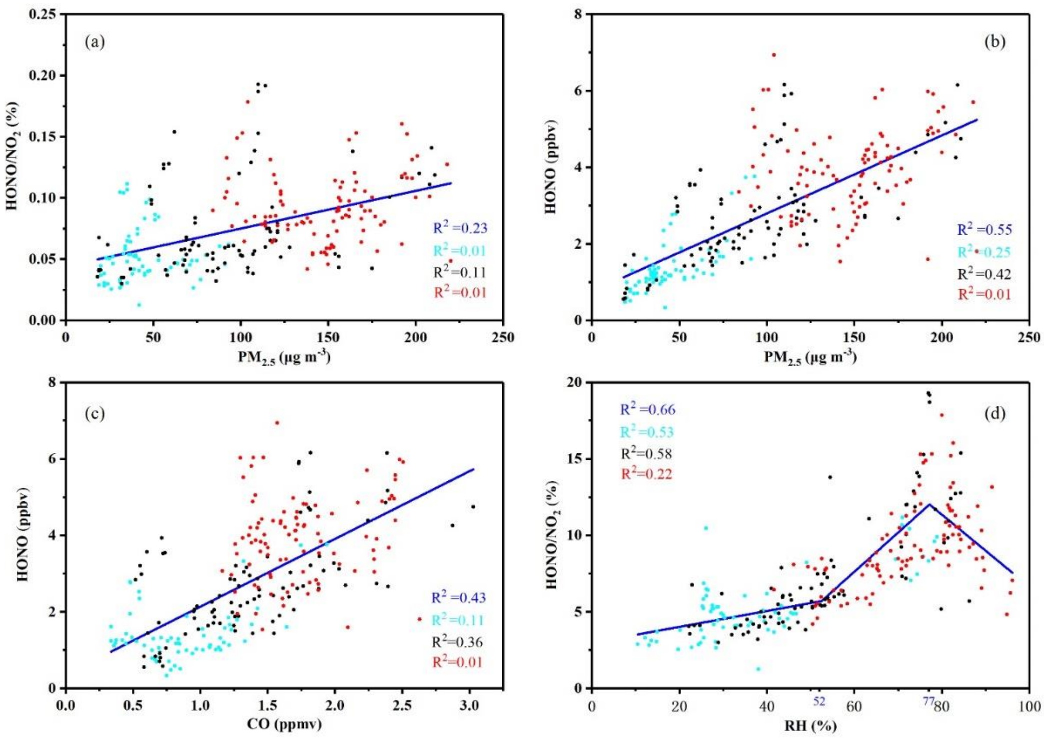
55. Fig. 4: Need error bars

With too many error bars in figures, it will make the figures look unclear. So the error bars of **Fig. 4**, **Fig. 5**, and **Fig. 8** were placed separately in the tables of the supplement (**Table S2**, **Table S3**, and **Table S4**).

56. Fig. 6: A space after HONO

We have modified the figure in the revised text **Fig. 7**.

Fig. 7. Nighttime correlation studies between $PM_{2.5}$ and $HONO/NO_2$, $PM_{2.5}$ and HONO, CO and HONO, RH and $HONO/NO_2$ during the entire measurement period, CD, PD, and SPD periods. The blue represented the full measurement period; the light blue represented CD period; the black represented PD period; the red represented SPD period.



Characteristics, sources, and reactions of nitrous acid during winter at an urban site in the Central Plains Economic Region in China

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Supplement:

1. This AIM method and its details.

HONO was hygroscopically grown in the parallel plate denuder and collected as an aqueous solution in a cyclone assembly. The aqueous sample aliquots from both channels were transported to the ion chromatographic systems housed inside a ground container for hourly semicontinuous online analysis of HONO. The ion chromatographic system was calibrated for NO_2^- using mixed anion standard solutions of NO_2^-.

2. The concentration of OH radicals was calculated with the formulas of NO_2, O_3, and J_{O^1D}。

$$[\mathrm{OH}] = \frac{k_{\mathrm{HO_2+NO}}\tau_{\mathrm{HC}}[\mathrm{NO_2}]F_J}{k_{\mathrm{NO+O_3}}} \times \sqrt{\frac{\alpha}{k_{HO_2+HO_2}[O_3]}} \times J(O^1D),$$

where [OH] represents the concentration of OH radicals, $k_{\mathrm{HO_2+NO}} = 8.56 \times 10^{-12}$ cm^3 s^{-1}, $\tau_{HC} = 0.3$ s, [NO_2] represents the NO_2 concentration, $F_J = 2$ s$^{-0.5}$, $k_{\mathrm{NO+O_3}} = 1.82 \times 10^{-14}$ cm^3 s^{-1}, $\alpha = 0.075$, $k_{\mathrm{HO_2+HO_2}} = 8.56 \times 10^{-12}$ cm^3 s^{-1}, [O_3] represents the O_3 concentration, and $J(O^1D)$ represents the O^1D efficiency of photolysis.

Figure Captions:

Fig. S1. The correlation study between $HONO_{correct}$ and NO_2 in the nighttime.

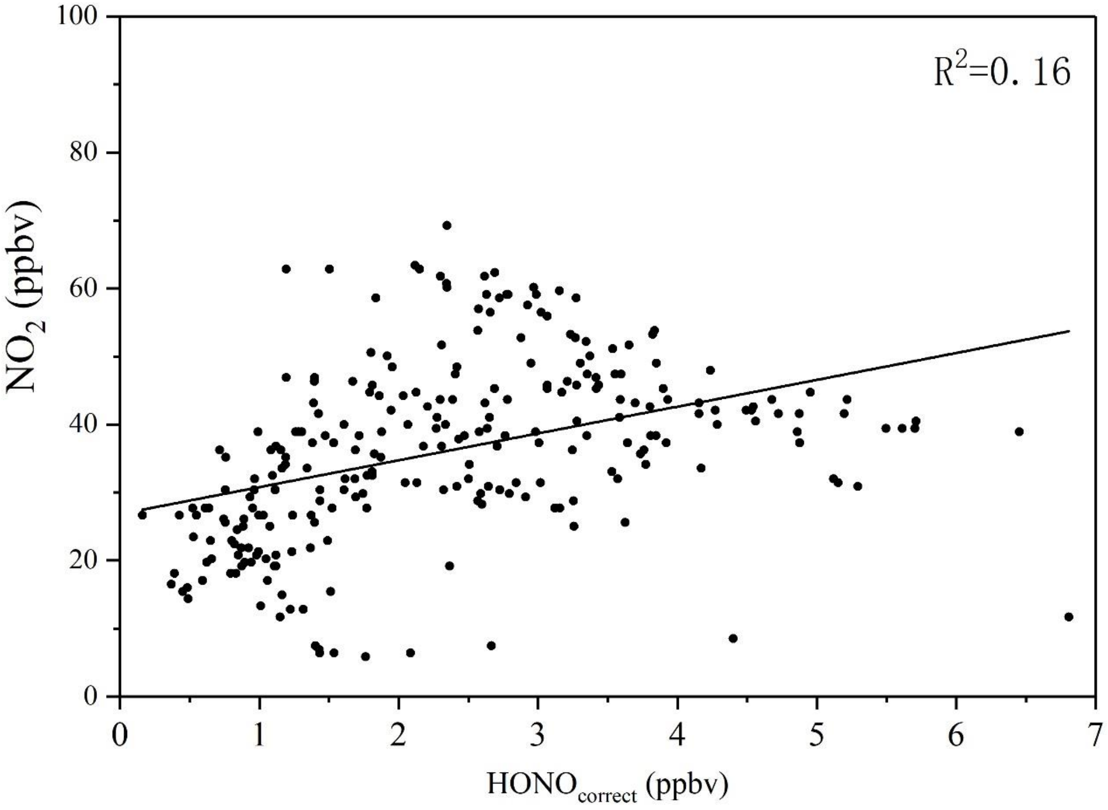

Fig. S1. The correlation study between $HONO_{correct}$ and NO_2 in the nighttime.

Table Captions:

Table S1. Measured species and performance of the instruments.

Table S2 The error bars of Fig. 4. (The units of all species except $HONO/NO_2$ and $HONO/NO_x$ are ppbv. The units of $HONO/NO_2$ and $HONO/NO_x$ are %.)

Table S3 The error bars of Fig. 5. (The units of all species except P_{OH+NO}^{net} are ppbv. The unit of P_{OH+NO}^{net} is ppbv/h.)

Table S4 The error bars of Fig. 8. (The units of all species except $HONO_{correct}/NO_2$ are ppbv. The unit of $HONO_{correct}/NO_2$ is %.)

Table S1. Measured species and performance of the instruments.

Species	Measurement technique	Detection limit	Accuracy
$PM_{2.5}$	Tapered Element Oscillating Microbalance	1.5 μg m^{-3}	± 5%
HONO	Ion Chromatography	4 pptv	± 20%
CO	Absorbs Infrared Radiation	40 ppbv	± 5%
NO	Chemiluminescence	60 pptv	± 20%
NO_2	Chemiluminescence	300 pptv	± 20%
O_3	UV Photometry	0.5 ppbv	± 5%

The results came from instrument manufacturers.

Table S2-1 The error bars of Fig. 4. (The units of all species except $HONO/NO_2$ and $HONO/NO_x$ are ppbv. The units of $HONO/NO_2$ and $HONO/NO_x$ are %.)

	Local Time (hh:mm)									
Species-period	00:00	01:00	02:00	03:00	04:00	05:00	06:00	07:00	08:00	09:00
HONO-CD	1.7 ± 1.3	1.4 ± 0.6	1.3 ± 0.4	1.2 ± 0.3	1.2 ± 0.2	1.2 ± 0.2	1.4 ± 0.3	1.5 ± 0.6	1.7 ± 0.9	1.6 ± 0.9
HONO-PD	3.2 ± 1.5	3.1 ± 1.3	3 ± 1.1	3.3 ± 1.2	3.5 ± 1.3	3.5 ± 1.2	3.6 ± 1.1	3.3 ± 0.9	3.7 ± 1.6	4.1 ± 2.8
HONO-SPD	3.7 ± 0.9	4 ± 0.8	4.2 ± 0.6	4.4 ± 0.8	4.6 ± 1	4.6 ± 1.2	4.6 ± 1.5	4.4 ± 1.3	4.4 ± 1.1	5.7 ± 3
NO-CD	14.3 ± 17	9 ± 9.7	8.5 ± 12.7	10.1 ± 22.4	10.6 ± 21.1	21.9 ± 29	27.8 ± 33	40.1 ± 51	52.6 ± 79	55.5 ± 84
NO-PD	57.3 ± 48	62.7 ± 55.9	49.6 ± 49	44 ± 47.8	47 ± 48.7	46.6 ± 30	41.4 ± 34	44.7 ± 33	48.9 ± 35	53.7 ± 44
NO-SPD	79.4 ± 103	100.1 ± 118	128.3 ±133	129 ± 134	111 ± 119	117 ± 95	100 ± 94	88.4 ± 85	82.3 ± 70	85.4 ± 71
NO_2-CD	25.4 ± 8.2	25.6 ± 9.9	24.7 ± 10.5	22.9 ± 10.4	24 ± 11.4	20.7 ± 11	20.2 ± 9	23.6 ± 11	28.6 ± 18	28.6 ± 18
NO_2-PD	41.1 ± 10	40.8 ± 11.2	39.7 ± 10.7	37.9 ± 7.1	36.6 ± 5.4	35.9 ± 5	33.8 ± 6	34.4 ± 6	33.2 ± 5	30.7 ± 6
NO_2-SPD	45.3 ± 9.5	43.5 ± 9.2	42.8 ± 8.8	42.1 ± 8.2	42.2 ± 8.1	41 ± 7.1	40.6 ± 6.9	40.7 ± 6	40.1 ± 6	39.2 ± 7
O_3-CD	14.2 ± 10	13.6 ± 10.4	14.2 ± 10.1	14.9 ± 9.4	13.6 ± 9.1	11.7 ± 10	13.8 ± 10	12.9 ± 9	11.6 ± 8	12.1 ± 7
O_3-PD	6.6 ± 6.1	6.4 ± 5.2	7.1 ± 5.2	6.3 ± 3.3	4.7 ± 2.2	5.3 ± 3	7.7 ± 6.9	5.3 ± 2.8	5.5 ± 3	7.1 ± 4
O_3-SPD	7.8 ± 6.4	7.7 ± 6.2	7.3 ± 5	6 ± 2.9	5.3 ± 2.3	5 ± 2.1	5.6 ± 2.5	5.2 ± 2.2	5.6 ± 2.6	6 ± 2.6
$HONO/NO_2$-CD	3.8 ± 1.5	4.4 ± 1	4.4 ± 1.1	4.9 ± 1	5.1 ± 0.8	8.3 ± 6	6.9 ± 2.1	6.2 ± 1.4	5.1 ± 0.8	4.3 ± 1.1
$HONO/NO_2$-PD	8 ± 3.6	7.8 ± 3.4	8 ± 3.3	9 ± 3.7	10 ± 4.5	10.1 ± 4	11.2 ± 4.6	10.3 ± 4	12.1 ± 7	14.3 ± 11
$HONO/NO_2$-SPD	8.3 ± 1.9	9.3 ± 1.4	10 ± 1.5	10.7 ± 1.9	11 ± 2.2	11.3 ± 3	11.5 ± 3.9	10.9 ± 3	11.1 ± 2	15 ± 8.3
$HONO/NO_x$-CD	2.7 ± 1.4	3.7 ± 1.5	4.2 ± 1.4	4.9 ± 1.1	4.9 ± 1	5.3 ± 2.5	5.1 ± 2.9	4.5 ± 2.4	3.6 ± 1.5	2.8 ± 1.4
$HONO/NO_x$-PD	4.4 ± 1.4	4.3 ± 1.7	4.6 ± 1.5	5.3 ± 1.3	5.3 ± 1	5.3 ± 1.1	6.6 ± 2.7	5.9 ± 2.3	6.5 ± 3.8	6.6 ± 4.3
$HONO/NO_x$-SPD	5.1 ± 2	5.3 ± 2.4	5.4 ± 3.4	5.8 ± 3.9	6.1 ± 3.9	5.7 ± 3.7	5.9 ± 3.6	5.7 ± 3	5.8 ± 2.9	6.7 ± 3.1

Table S2-2 The error bars of Fig. 4. (The units of all species except $HONO/NO_2$ and $HONO/NO_x$ are ppbv. The units of $HONO/NO_2$ and $HONO/NO_x$ are %.)

	Local Time (hh:mm)									
Species-period	10:00	11:00	12:00	13:00	14:00	15:00	16:00	17:00	18:00	19:00
HONO-CD	1.1 ± 0.6	0.6 ± 0.3	0.5 ± 0.3	0.6 ± 0.4	0.6 ± 0.5	0.7 ± 0.5	0.6 ± 0.5	0.7 ± 0.4	1 ± 0.5	1.2 ± 0.5
HONO-PD	2.9 ± 1.9	1.9 ± 1.3	1.3 ± 0.7	1 ± 0.3	0.9 ± 0.3	0.9 ± 0.3	0.9 ± 0.3	1.1 ± 0.4	1.4 ± 0.3	1.7 ± 0.3
HONO-SPD	6.9 ± 4.3	5.2 ± 3.8	3 ± 1.3	2.1 ± 0.7	1.8 ± 0.7	1.7 ± 0.6	1.8 ± 0.7	2 ± 0.5	2.7 ± 0.7	2.8 ± 0.8
NO-CD	43.9 ± 69.8	27.9 ± 40.8	14.9 ± 17.1	10.3 ± 7.8	7.3 ± 3	6 ± 4.5	6.4 ± 5.6	3.6 ± 3.4	2.6 ± 3.2	5.9 ± 7.7
NO-PD	49.3 ± 45.2	30 ± 26.2	21 ± 20.7	12.7 ± 14.7	9.4 ± 12.3	8.4 ± 9.5	5.7 ± 4.7	6.3 ± 6.8	9 ± 9	10 ± 10.3
NO-SPD	90.8 ± 73.4	79.3 ± 69.3	57.1 ± 52.3	34.8 ± 36.4	24.5 ± 28.7	19 ± 24.7	15 ± 18.8	11.8 ± 11	11.8 ± 7.9	22.4 ± 21
NO_2-CD	26.8 ± 15.7	22.7 ± 9.2	17.6 ± 7.1	17.1 ± 9	19.6 ± 9.6	21 ± 10.7	20.5 ± 9	21.4 ± 9	26 ± 12.5	30 ± 13.7
NO_2-PD	30 ± 6.9	28.8 ± 7.7	27.4 ± 9.6	24.8 ± 9.4	22.5 ± 10.6	25 ± 9.9	25.7 ± 9.3	27.1 ± 9	35 ± 8.7	36.2 ± 9.2
NO_2-SPD	39.8 ± 7.8	41.5 ± 8.3	42.3 ± 10.1	39.5 ± 12.6	38.5 ± 14.3	38 ± 14.7	38 ± 13.9	42 ± 15.4	45 ± 11.5	47 ± 10.8
O_3-CD	15.9 ± 8.8	19.5 ± 9.7	22.6 ± 8.3	25.5 ± 8.5	28.1 ± 9.1	29 ± 10.8	28 ± 10.8	29 ± 10.2	23.6 ± 10	17 ± 8.9
O_3-PD	9.6 ± 6.1	12.8 ± 6.2	18.7 ± 8.3	24.1 ± 8.4	28.2 ± 9.7	27 ± 10.8	28 ± 10.4	26 ± 10.5	17.4 ± 8.6	15 ± 11.6
O_3-SPD	6.3 ± 2.4	8.7 ± 4.5	12.8 ± 8.5	19.4 ± 12.9	24.1 ± 14.7	28 ± 16.6	29 ± 17.6	25 ± 16.1	17 ± 11.1	10.6 ± 9.7
$HONO/NO_2$-CD	4.1 ± 2.3	3.1 ± 1.9	3.3 ± 1.9	3.3 ± 1.3	3.1 ± 1.3	3.1 ± 1.3	2.9 ± 1.4	3.1 ± 1.4	3.9 ± 1.4	4.5 ± 2.2
$HONO/NO_2$-PD	9.4 ± 5.6	6.2 ± 3	4.7 ± 1.5	4.2 ± 1.2	4.7 ± 2.2	3.9 ± 0.7	3.7 ± 0.4	4.1 ± 1.2	4.3 ± 0.9	5 ± 1.5
$HONO/NO_2$-SPD	18.9 ± 13.7	13.7 ± 12	7.3 ± 3.5	5.6 ± 2.6	4.9 ± 2.1	4.8 ± 2.4	4.9 ± 1.6	5 ± 1	6.3 ± 1.8	6.2 ± 1.5
$HONO/NO_x$-CD	2.9 ± 2.1	2.2 ± 1.5	2.4 ± 1.5	2.5 ± 1.1	2.5 ± 1	2.6 ± 0.9	2.5 ± 0.9	2.8 ± 1	3.7 ± 1.1	4.1 ± 1.9
$HONO/NO_x$-PD	4.8 ± 2.4	3.8 ± 1.3	3.5 ± 1.2	3.5 ± 1.5	4 ± 2.1	3.4 ± 0.9	3.3 ± 0.5	3.7 ± 1.2	3.8 ± 0.7	4.3 ± 1.5
$HONO/NO_x$-SPD	8.2 ± 5.8	6.9 ± 5.7	4.3 ± 2	4 ± 2	3.8 ± 1.6	3.9 ± 1.9	4.3 ± 1.6	4.5 ± 1.2	5.5 ± 1.5	4.9 ± 1.3

Table S2-3 The error bars of Fig. 4. (The units of all species except $HONO/NO_2$ and $HONO/NO_x$ are ppbv. The units of $HONO/NO_2$ and $HONO/NO_x$ are %.)

	Local Time (hh:mm)			
Species-period	20:00	21:00	22:00	23:00
HONO-CD	1.3 ± 0.6	1.6 ± 0.9	2 ± 0.9	2.1 ± 0.9
HONO-PD	1.7 ± 0.7	1.8 ± 0.8	2 ± 0.9	2.1 ± 0.9
HONO-SPD	3.1 ± 0.9	3.2 ± 0.9	3.7 ± 0.8	4.6 ± 1.2
NO-CD	11.1 ± 16.9	14.5 ± 22.5	35.5 ± 68.9	50.8 ± 99.2
NO-PD	15 ± 14.1	15.3 ± 14.7	27.4 ± 28.5	33.9 ± 28.9
NO-SPD	29.4 ± 24.2	37.3 ± 26.6	38.5 ± 23.1	51.4 ± 31.4
NO_2-CD	31 ± 13.8	30.3 ± 14.5	31.6 ± 13.6	31 ± 14.3
NO_2-PD	37.3 ± 10.5	38.5 ± 13.9	38.3 ± 13.5	37.1 ± 13.2
NO_2-SPD	44.5 ± 11	43.5 ± 11.5	43.5 ± 11.1	42.1 ± 13.1
O_3-CD	13.3 ± 10.1	14 ± 11	12.2 ± 8.7	12.7 ± 8.8
O_3-PD	13.7 ± 10.3	10.9 ± 8.5	10.9 ± 7.7	12.2 ± 10.4
O_3-SPD	9.9 ± 8.6	10.8 ± 9.2	9.7 ± 8.7	9.6 ± 9.6
$HONO/NO_2$-CD	4.6 ± 2.2	5.7 ± 2.6	6.5 ± 2.6	6.8 ± 2.7
$HONO/NO_2$-PD	4.7 ± 1.9	4.6 ± 1.2	4.9 ± 0.8	5.3 ± 0.8
$HONO/NO_2$-SPD	7 ± 1.5	7.5 ± 1.4	8.9 ± 2.3	9.4 ± 2.4
$HONO/NO_x$-CD	4 ± 1.9	4.8 ± 2.2	4.9 ± 2.8	5 ± 3
$HONO/NO_x$-PD	3.9 ± 2.1	3.9 ± 1.3	3.8 ± 1	3.8 ± 0.9
$HONO/NO_x$-SPD	5.1 ± 1.5	5.2 ± 2	5.8 ± 2	5 ± 1.4

Table S3-1 The error bars of Fig. 5. (The units of all species except P_{OH+NO}^{net} are ppbv. The unit of P_{OH+NO}^{net} is ppbv/h.)

	Local Time (hh:mm)									
Species-period	19:00	20:00	21:00	22:00	23:00	00:00	01:00	02:00	03:00	04:00
P_{OH+NO}^{net}-CD	0.04 ± 0.06	0.08 ± 0.12	0.11 ± 0.17	0.33 ± 0.54	0.47 ± 0.79	0.12 ± 0.13	0.07 ± 0.08	0.03 ± 0.03	0.01 ± 0.1	0.02 ± 0.1
HONO-CD	1.18 ± 0.48	1.32 ± 0.62	1.62 ± 0.9	2.02 ± 0.94	2.09 ± 0.9	1.67 ± 1.34	1.43 ± 0.63	1.26 ± 0.44	1.2 ± 0.3	1.2 ± 0.22
NO-CD	5.4 ± 6.5	10.2 ± 14.4	13.3 ± 19.2	38.2 ± 62.2	54.9 ± 89.7	15 ± 14.8	8.8 ± 8.6	3.7 ± 4.2	1.5 ± 2.3	2.5 ± 2.6
P_{OH+NO}^{net}-HD	0.07 ± 0.07	0.1 ± 0.1	0.1 ± 0.1	0.19 ± 0.2	0.23 ± 0.2	0.4 ± 0.34	0.44 ± 0.4	0.34 ± 0.35	0.3 ± 0.34	0.3 ± 0.34
HONO-HD	1.7 ± 0.27	1.71 ± 0.68	1.82 ± 0.78	1.98 ± 0.89	2.06 ± 0.93	3.21 ± 1.54	3.05 ± 1.27	3.01 ± 1.08	3.3 ± 1.17	3.5 ± 1.34
NO-HD	8.5 ± 8.4	12.2 ± 11.5	12.5 ± 12	22.4 ± 23.3	27.7 ± 23.6	46.8 ± 39.5	51.2 ± 45.6	40.5 ± 40	35.9 ± 39	38 ± 39.7
P_{OH+NO}^{net}-SHD	0.15 ± 0.15	0.2 ± 0.17	0.25 ± 0.18	0.26 ± 0.16	0.35 ± 0.23	0.55 ± 0.75	0.7 ± 0.85	0.9 ± 0.96	0.9 ± 1.0	0.8 ± 0.86
HONO-SHD	2.8 ± 0.8	3.1 ± 0.9	3.2 ± 0.9	3.7 ± 0.8	4.6 ± 1.2	3.7 ± 0.9	4 ± 0.8	4.2 ± 0.6	4.4 ± 0.8	4.6 ± 1
NO-SHD	18 ± 17	24 ± 20	30 ± 21	31 ± 19	42 ± 25	64 ± 84	81 ± 96	104 ± 108	105 ± 110	90 ± 97

Table S3-2 The error bars of Fig. 5. (The units of all species except P_{OH+NO}^{net} are ppbv. The unit of P_{OH+NO}^{net} is ppbv/h.)

	Local Time (hh:mm)	
Species-period	05:00	06:00
P_{OH+NO}^{net}-CD	0.12 ± 0.18	0.17 ± 0.22
HONO-CD	1.25 ± 0.21	1.36 ± 0.35
NO-CD	13.7 ± 20.9	19.5 ± 25.1
P_{OH+NO}^{net}-HD	0.32 ± 0.22	0.28 ± 0.25
HONO-HD	3.5 ± 1.16	3.56 ± 1.09
NO-HD	38 ± 25.2	33.8 ± 28.5
P_{OH+NO}^{net}-SHD	0.82 ± 0.87	0.7 ± 0.68
HONO-SHD	4.6 ± 1.2	4.6 ± 1.5
NO-SHD	95.6 ± 99	81.8 ± 77.1

Table S4-1 The error bars of Fig. 8. (The units of all species except $HONO_{correct}/NO_2$ are ppbv. The unit of $HONO_{correct}/NO_2$ is %.)

	Local Time (hh:mm)									
Species-period	19:00	20:00	21:00	22:00	23:00	00:00	01:00	02:00	03:00	04:00
$HONO_{correct}$-CD	1.0 ± 0.4	1.1 ± 0.6	1.4 ± 0.8	1.6 ± 0.7	1.6 ± 0.6	1.4 ± 1.4	1.2 ± 0.7	1.1 ± 0.5	1.1 ± 0.4	1.1 ± 0.2
NO_2-CD	30 ± 15	31 ± 15	30 ± 15	34 ± 15	34 ± 15	25 ± 9	24 ± 8	22 ± 8	20 ± 8	20 ± 8
$HONO_{correct}/NO_2$-CD	3.7 ± 2.2	3.9 ± 2.2	4.9 ± 2.6	5.5 ± 2.7	5.7 ± 2.9	11 ± 18.2	8.9 ± 12	8.6 ± 10.8	8.5 ± 9.7	7.7 ± 7.4
$HONO_{correct}$-HD	1.4 ± 0.3	1.4 ± 0.7	1.5 ± 0.7	1.6 ± 0.8	1.7 ± 0.8	2.7 ± 1.3	2.5 ± 1	2.5 ± 0.8	2.9 ± 0.9	3.1 ± 1.1
NO2-HD	36 ± 9	37 ± 10	39 ± 14	38 ± 13	37 ± 13	41 ± 10	41 ± 11	40 ± 11	38 ± 7	37 ± 5
$HONO_{correct}/NO_2$-HD	4.2 ± 1.5	3.8 ± 2	3.8 ± 1.2	4 ± 0.8	4.4 ± 0.7	6.7 ± 3.1	6.5 ± 2.8	6.7 ± 2.8	7.8 ± 3.1	8.7 ± 3.8
$HONO_{correct}$-SHD	2.4 ± 0.6	2.6 ± 0.7	2.7 ± 0.7	3.2 ± 0.7	4.1 ± 1.3	3.1 ± 0.8	3.3 ± 0.6	3.4 ± 0.7	3.6 ± 1	3.9 ± 1.1
NO_2-SHD	47 ± 11	44 ± 11	43 ± 11	44 ± 11	42 ± 13	45 ± 9	43 ± 9	43 ± 9	42 ± 8	42 ± 8
$HONO_{correct}/NO_2$-SHD	5.4 ± 1.4	6.1 ± 1.4	6.5 ± 1.4	7.8 ± 2.2	14.4 ± 16.7	7 ± 1.9	7.8 ± 1.6	8.1 ± 2.2	8.8 ± 2.8	9.3 ± 2.9

Table S4-2 The error bars of Fig. 8. (The units of all species except $HONO_{correct}/NO_2$ are ppbv. The unit of $HONO_{correct}/NO_2$ is %.)

	Local Time (hh:mm)	
Species-period	05:00	06:00
$HONO_{correct}$-CD	1.0 ± 0.4	1.1 ± 0.6
NO_2-CD	30 ± 15	31 ± 15
$HONO_{correct}/NO_2$-CD	3.7 ± 2.2	3.9 ± 2.2
$HONO_{correct}$-HD	1.4 ± 0.3	1.4 ± 0.7
NO_2-HD	36 ± 9	37 ± 10
$HONO_{correct}/NO_2$-HD	4.2 ± 1.5	3.8 ± 2
$HONO_{correct}$-SHD	2.4 ± 0.6	2.6 ± 0.7
NO_2-SHD	47 ± 11	44 ± 11
$HONO_{correct}/NO_2$-SHD	5.4 ± 1.4	6.1 ± 1.4

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