

#### *AUTHOR RESPONSES IN BLUE ITALIC TEXT*

The manuscript “Exploration of the atmospheric chemistry of nitrous acid in a coastal city of southeastern China: Results from measurements across four seasons” by Baoye Hu et al. reports year-long observations of HONO together with gaseous, particulate, and meteorological parameters which are relevant for investigating HONO sources. The manuscript adds valuable information on HONO concentration level and its temporal variation under coastal condition. Discussions on the HONO sources and on the HONO impacts on OH radical production is however in line with the current understanding. I would recommend the publication if my following comments are well addressed.

*Response: Thanks for your valuable comments and positive feedback. We have corrected this manuscript according to your suggestion. Below are the point-to-point responses to general and specific comments.*

#### **General comments**

There are plenty of published papers describing HONO measurements, most of which are also using the similar methodology to investigate HONO sources and draw similar conclusion that daytime HONO is mainly originated from photolysis of nitrates. In order to make the manuscript more valuable to the community, I would suggest the authors put more efforts on summarizing the findings on HONO production in different environments (e.g., inland or coastal, downtown or suburban or rural, seasons, RH levels, NO<sub>3</sub> levels, etc.) and compare those with this work. I think the comprehensive data set shown in the manuscript would well support the comparison. The current comparisons listed in Table 1 and Table 3 are too general and not quite informative compared to those already shown in many other publications.

*Response: We would like to thank the reviewer’s valuable comments and effort in reviewing the manuscript. We have summarized the findings on HONO production in different environments and compared those with this work. Since many studies did not measure NO<sub>3</sub> concentration, we used the most frequently measured NO and NO<sub>2</sub> concentration instead. As shown in Table 1, the HONO concentration measured at this site was comparable to those measured at other suburban sites (Liu et al., 2019; Xu et al., 2015; Nie et al., 2015; Park et al., 2004), was obvious lower than those measured at urban sites and industrial site (Li et al., 2018; Yu et al., 2009; Hou et al., 2016; Qin et al., 2009; Wang et al., 2013; Shi et al., 2020; Spataro et al., 2013; Huang et al., 2017; Wang et al., 2017), and was obvious higher than those measured at marine background (Wen et al., 2019), Marine boundary layer (Ye et al., 2016), and coastal remote (Meusel et al., 2016). Besides, this table was put in “Supplementary Material” due to its length.*

Table 1. Comparison of HONO concentrations and related parameters at this site with other regions.

Site	Country	Type	Seasons	RH (%)	T (°C)	NO/NO <sub>2</sub>	HONO (ppb)	Reference
Jinan	China	Urban	Annual	51.42	16.07	15.38/27.92	1.15 ± 1.07	(Li et al., 2018)
			Spring	56.67	16.77	11.33/29.67	1.16 ± 0.90	
			Summer	38.67	26.67	5.67/17.33	1.12 ± 0.93	
			Autumn	53.00	16.33	13.00/23.67	0.78 ± 0.60	
			Winter	59.67	3.00	31.17/36.33	1.71 ± 1.62	
Kathmandu	Nepal	Urban	Winter	—	—	3.16/14.14	1.55	(Yu et al., 2009)
Beijing	China	Urban	Severe haze	—	—	29.35/48.1	1.95	(Hou et al., 2016)
			Clean	—	—	5.2/18.85	0.72	
Guangzhou	China	Urban	Summer	77	31.2	—/30.3	~2.8	(Qin et al., 2009)
Shanghai	China	Urban	Annual	—	—	—/18.78	0.92 ± 0.57	(Wang et al., 2013)
Changzhou	China	Urban	Spring	53.7 ± 19.8	18.7 ± 4.8	8.2/22.9	1.55 ± 1.21	(Shi et al., 2020)
Beijing	China	Urban	Summer	56.79	28.27	6.44/31.70	1.45 ± 0.58	(Spataro et al., 2013)
			Winter	26.02	3.51	25.90/38.76	1.04 ± 0.73	
Xi'an	China	Urban	Summer	—	—	—/20.9	1.12 ± 0.97	(Huang et al., 2017)
Beijing	China	Urban	Annual	43.34	16.01	—/25.60	1.44 ± 1.33	(Wang et al., 2017)
			Spring	34.73	18.44	—/25.97	1.05 ± 0.95	
			Summer	55.30	28.11	—/19.21	1.38 ± 0.90	
			autumn	51.11	17.33	—/32.91	2.27 ± 1.82	
			winter	30.48	-3.57	—/19.96	1.05 ± 0.89	
Nanjing	China	Industrial	Winter	68	6.1	7.97/23.9	1.32 ± 0.92	(Zheng et al., 2020)
Touji Island	China	Marine background	Autumn	74	14.2	0.5/5.3	0.20 ± 0.20	(Wen et al., 2019)
The North Atlantic Ocean	—	Marine boundary layer	5 July 2013	—	—	—	0.0113 ± 0.0016	(Ye et al., 2016)
			8 July 2013	—	—	—	0.0088 ± 0.0023	

Cyprus	Mediterranean Sea	Coastal remote	summer	—	18-28	0.02/0.14	0.035 ± 0.025	(Meusel et al., 2016)
Kwangju	Korea	Suburban	Autumn	74.55	15.08	—	0.67 ± 0.60	(Park et al., 2004)
Nanjing	China	Suburban	Annual	72	17.00	5.7/16.4	0.69 ± 0.58	(Liu et al., 2019)
			Spring	73	17.67	2.35/15.15	0.68 ± 0.48	
			Summer	77	28.00	1.2/10.1	0.45 ± 0.37	
			Autumn	72	18.11	5.25/16.15	0.66 ± 0.53	
			Winter	66	4.33	15.58/25.75	1.04 ± 0.75	
Hongkong	China	Suburban	Annual	73	25	10.7/21.7	0.71	(Xu et al., 2015)
			Spring	75	28	5.5/15.5	0.35	
			Summer	71	32	8/19.8	0.65	
			Autumn	67	23	10.1/26.8	0.93	
			Winter	78	17	19.3/24.7	0.91	
Western Yangtze River delta	China	Suburban	Spring	—	—	—	0.76 ± 0.79	(Nie et al., 2015)
Xiamen	China	Suburban	Annual	78.35	22.95	5.80/14.99	0.54 ± 0.47	This work
			Spring	84.21	16.59	8.47/18.10	0.62 ± 0.58	
			Summer	84.12	30.00	4.79/13.39	0.61 ± 0.39	
			Autumn	69.55	24.02	2.18/12.88	0.41 ± 0.30	
			Winter	78.13	18.41	8.86/17.03	0.54 ± 0.47	

Note: “—” means no data found in the corresponding reference.

## Specific comments

**Line 93 – 96, Page 4:** The author should make a clear description that the time needed for the sampling period and the later IC analysis on the MARGA system. When synchronize high time resolution data (i.e., HONO, NO<sub>x</sub>, J values) to MARGA data, it should be done exactly for the MARGA sampling period. This should also be clearly described.

*Response: Thanks for your valuable suggestion. We have made a clear description that the time needed for the sampling period and the later IC analysis on the MARGA system. The inorganic composition of PM<sub>2.5</sub> aerosols (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>) and concentrations of gases (HONO, HNO<sub>3</sub>, HCl, SO<sub>2</sub>, NH<sub>3</sub>) were determined using a Monitor for Aerosols and Gases in ambient Air (MARGA, Model ADI 2080, Applikon Analytical B.V., the Netherlands) consists of two identical sample boxes and one analytical box. Ambient air was drawn at the flow rate of 1 m<sup>3</sup>·h<sup>-1</sup> into the sample box by a PM<sub>2.5</sub> cyclone (Teflon coated inlet, URG-2000-30ENB). Sample air was first drawn through the Wet Rotating Denuder (WRD) where water-soluble gases diffused to the absorption solution, then particles were collected in a Steam Jet Aerosol Collector (SJAC). Absorption solutions were drawn from the WRD and the SJAC to syringes (25 ml) in the analytical box. Each hour after the syringes had been filled, samples were injected to Metrohm anion (250 µl loop) and cation (500 µl loop) chromatographs with the internal standard (LiBr) for 15 minutes (Makkonen et al., 2012). Specific descriptions of the SJAC can be found in previous reports (Slanina et al., 2001; Wyers et al., 1993). Therefore, the times needed for the sampling period and the latter IC analysis on the MARGA system are a full hour and 15 minutes, respectively. The value measured in this hour is actually the concentration sampled in the previous hour, so the time corresponding to the sampling is matched with other instrument parameters (i.e., HONO, NO<sub>x</sub>, J values).*

**Line 110, Page 4:** I would recommend the Section 3.1 focus on reporting the measurement results, discussions on HONO sources and OH production by HONO photolysis can be moved to the following specific sections.

*Response: Thanks for your careful working. This way of writing is more logical. Some conclusions based on observations are more convincing and avoid repeating the results. The more specific discussion on HONO source focused on the Sec 3.2, 3.3, 3.4, and OH production by HONO photolysis focused on the Sec 3.6.*

**Line 111 – 115, Page 4:** As mentioned in the General Comments, I suggest to make the comparison in terms of environmental conditions.

*Response: Thanks for your valuable suggestion. The comparison in terms of environmental conditions has been made as suggested in revised manuscript.*

**Line 128 – 131, Page 5:** How about the sea salt concentration observed in this study? The argument is based on the assumption that HONO is mainly formed by photolysis of sea salt, which could not be confirmed at this moment.

*Response: Thanks for your valuable suggestion. The sea salt concentration was 2.91 µg·m<sup>-3</sup> during the day and 2.73 µg·m<sup>-3</sup> during the night. And the sea salt concentration during the day is significantly higher than that during the night. This assumption was based on the reference that nitrate photolysis of sea salt particulate acts as a significant source of HONO (Kasibhatla et al., 2018).*

**Line 134 – 135, Page 5:** It would be helpful to confirm this by using the measured J, NO<sub>x</sub>, RH, etc.

*Response: This conclusion had been confirmed by J and NO<sub>x</sub> in the next paragraph and by RH in Sec. 3.3.2.*

**Line 162 – 163, Page 6:** I could not follow the argument that higher HONO/NO<sub>x</sub> ratio indicate unknown daytime HONO sources. The authors should first describe what is “unknown” and if the observation could not be explained by the well accepted theory.

*Response: Thanks for your valuable suggestion. This conclusion was based on the reference that the large HONO/NO<sub>x</sub> observed at around noon revealed additional daytime source(s) of HONO(Xu et al., 2015;Liu et al., 2019). If the HONO sources during the daytime are consistent with those at night, the minimum HONO/NO<sub>x</sub> ratio should occur at noon due to the intense photochemical loss of HONO. Therefore, there must be additional sources of HONO during daytime. “Unknown” has been changed into “additional” in order to avoid misunderstanding.*

**Line 184, Page 6:** How is the duration of air masses been determined?

*Response: Thanks for your careful working. The meaning of the text is that the air mass meets the conditions (1), (3), (4) and (5), and the duration of the air mass cannot exceed 2 h (Liu et al., 2019;Xu et al., 2015), which was based on the following two reasons. Firstly, fresh air mass should be short time. An Air mass with high NO<sub>x</sub> lasting long could not be a local fresh air mass, but an aged air mass transporting from high NO<sub>x</sub> region, such as city region. Secondly, if the duration of air mass was too long, the HONO observed was easily affected by secondary production, which would overestimate vehicle emission.*

**Line 242, Page 8 – Line 243 Page 9:** Seeing from the summer plot in Fig. 4, most blue points lie in values below 0.03 when RH is above 90%. It looks to me that the trend of orange line is biased by only few data points which have high HONO to NO<sub>2</sub> ratio.

*Response: Thanks for your careful working. It is true that most blue points lie in values below 0.03 when RH is above 90%. However, top-five HONO<sub>corr</sub>/NO<sub>2</sub> ratios would reduce the influence of those circumstances such as advection, the time of the night, and the surface density(Li et al., 2012;Stutz et al., 2004).Therefore, top-five HONO<sub>corr</sub>/NO<sub>2</sub> ratios were applied to replace all ratios.*

**Line 312, Page 11:** Should be “Eq. (7)” instead of “Eq. (6)”.

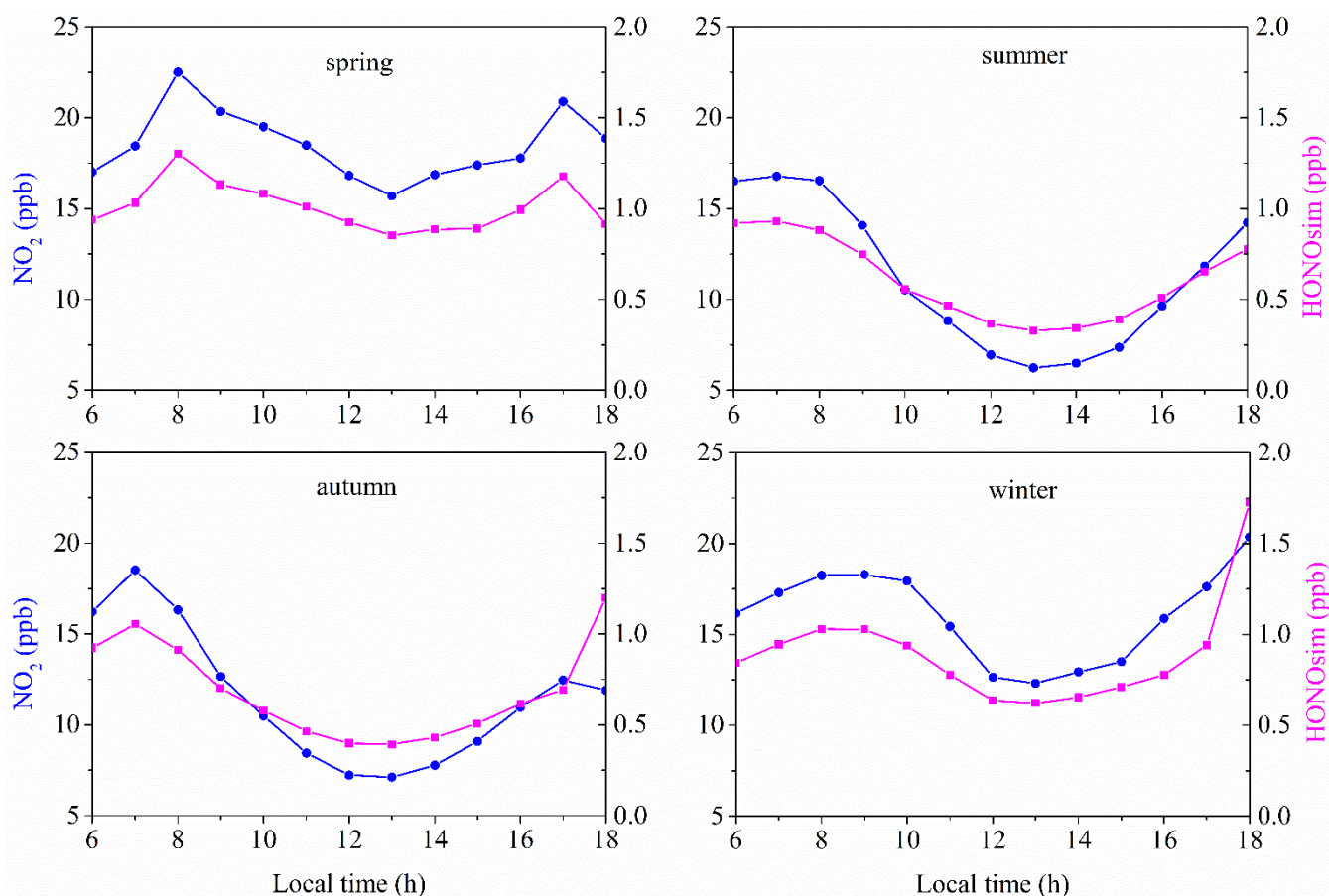
*Response: Revised.*

**Section 3.5, Page 13 – 14:** First of all, there are various assumptions on HONO production pathways been made in the previous sections. I would be better to provide a full picture on how large of the each contribution to the HONO formation. Secondly, under the title “Parameterization”, the reader could not even find a formula used for predicting HONO production or concentration. Moreover, why Eq. (10) is suitable in other place than in this work? Would the parameterization described in this work more reasonable and can be better used in the future?

*Response: Thanks for your valuable suggestion. Firstly, the contribution of each HONO production pathways has been specifically displayed in Section 3.4.2. Secondly, although the formula used for predicting HONO concentration in this study had not been explicitly given, we had described in the manuscript what kind of method can be used to improve parameterize., such as “Therefore, we used slopes of  $2.60 \times 10^{-2}$ ,  $2.06 \times 10^{-2}$ , and  $1.59 \times 10^{-2}$  to parameterize the HONO concentrations in summer, autumn, and winter, respectively. As for spring, though only a weak correlation between HONO and NO<sub>x</sub> was found, the majority of the HONO/NO<sub>x</sub> ratios fluctuated round a slope of 0.02 because concentrations of NO<sub>x</sub> greater than 60 ppb only accounted for 8.83% of the data. Therefore, a slope of 0.02 was applied in spring to parameterize the HONO concentration.”, and*



“Therefore, we take the photolysis of nitrate into the spring, summer, and autumn calculations, but we use the reaction of NO and OH in the calculations for winter. In this way, the daytime simulation results are significantly improved”. Eq. (10) ( $[HONO] = 0.01 \times [NO_2] \times J(NO_2)/J(HONO)$ ) can be regarded as a combination of  $[NO_2]$  with  $J(NO_2)/J(HONO)$ .  $J(NO_2)/J(HONO)$  kept relatively constant (5.34~5.69) in the daytime in four seasons. Therefore, diurnal variation of  $[HONO]$  simulated by Eq. (10) depended on  $[NO_2]$  (Figure 1). This formula is only suitable for regions where the diurnal variation of  $[NO_2]$  is consistent with that of  $[HONO]$ . The parameterization described in this work was more reasonable and can be better used in the future in such coastal sites. Whether it is applicable to other types of sites needs to be further verified in the future. Figure 1 has been added in “Supplementary Material” named as Figure S5.



**Figure 1. Diurnal variation of  $NO_2$  concentration and HONO concentration simulated by Eq. (10)**

**Section 3.6, Page 14 – 15:** I think the authors should make a clear statement that they are evaluation the primary production of OH radical. As shown in many publications investigating HOx budgets, the production of OH during daytime is mainly by  $HO_2 / RO_2 + NO$  reaction.

*Response: Thanks for your valuable suggestion. The reaction of  $RO_2/HO_2$  with NO is indeed an important source of OH radicals (Wang et al., 2018). Besides,  $O_3$  photolysis, HONO photolysis, hydrogen peroxide photolysis and the ozonolysis of alkenes are main source of OH radicals. It is a pity that we do not have the supporting measurements to do this accurately except for  $O_3$  and HONO. Therefore, we added a clear statement about the primary productions of OH radical in the manuscript like this: In addition to the two primary production of OH radicals mentioned above, there are the reaction of organic and hydro peroxy radicals ( $RO_2$  and  $HO_2$ ) with NO,  $O_3$  photolysis, HONO photolysis, hydrogen peroxide photolysis and the ozonolysis of alkenes (Hofzumahaus et al., 2009; Gligorovski et al., 2015; Wang et al., 2018).*

## References

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