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 Hu et al. provides observations and analysis of compounds important for improving understanding of tropospheric chemistry. The topic is important to many readers, and this study is closely related to a large number of papers that try to understand atmospheric HONO abundance and its impact on oxidants. The writing is clear, and the observations are sufficiently unique and comprehensive to provide new insights. Regrettably, the utility of the manuscript is compromised by the omission of many important experimental details, such that the context and relevance of the measurements reported here cannot be ascertained. Additionally, there are several analyses that are incomplete or difficult to understand. Consequently, I cannot recommend this paper for publication. I do hope that these measurements will receive further examination and that a paper will be written that considers some of the suggestions below.

Response: Thanks for your positive feedback for the whole manuscript and valuable comments for some details. We have tried our best to improve the quality of this manuscript. Many experimental details have been added in the corresponding section to ascertain the context and relevance of the measurements. Several analyses have been improved to be complete and easy to understand.

The methodology section is far too brief, and many critical details are absent. The reference (Duan et al, 2018) that describes the HONO instrument notes the importance of characterizing HONO transmission and production in inlets. There is no mention of any of the sampling inlets. NO_2 readily converts to HONO on surfaces, but there is no way to assess the importance of this artifact without a thorough description of inlet length, material, flow, etc. Are filters used on the IBBCEAS to remove ambient aerosol, as in Min et al., 2016? If so, how often are they changed? What are the uncertainties for the aerosol and NO measurements? It would be helpful to show how the IBBCEAS and TEI NO_2 measurements compare. Line 105 says the TEI "might actually include other active nitrogen compounds". Did it? This assertion should be tested, or at least referenced.

Response: Thanks for your careful and precise working. The methodology section has been improved a lot as follows: The atmospheric concentrations of both HONO and NO_2 were determined using IBBCEAS, which has previously been widely applied to such measurements (Tang et al., 2019;Duan et al., 2018;Min et al., 2016). The custom-built IBBCEAS instrument from the Anhui Institute of Optics and Fine Mechanic (AIOFM), Chinese Academy of Sciences, has been described in detail in previous study (Duan et al., 2018). Therefore, only a brief description is given here. Light was emitted by a single light-emitting diode (LED) with peak wavelength of 365 nm, full width at half maximum (FWHM) of 13 nm and was introduced into the resonant cavity, consisting of a pair of high-reflective (HR) mirrors with reflectivity of about 0.99983 at 368.2 nm, separated by 70 cm. The surface of the mirrors was purged by dry nitrogen at 0.1 Standard Liter per Minute (SLM), and the air flow was controlled by mass flow controller to prevent the surface of the mirror from being contaminated. The light transmitted through the cavity was received by an QE65000 spectrometer (Ocean Optics) through an optical fiber with 600 μm diameter and a 0.22 numerical aperture.

In order to avoid the drift of the center wavelength of the LED, the temperature of the LED was controlled to be approximately 25±0.01 ℃ by using a thermoelectric cooler unit. In order to prevent particulate matter from entering the cavity and reducing the effect of particulate matter on the effective absorption path, a 1 μm polytetrafluoroethylene (PTFE) filter membrane (Tisch Scientific) was used in the front end of the sampling port. In order to assure the quality of the data, the 1 μm PTFE filter membrane was usually replaced once every three days and the sampling tube was thoroughly cleaned with alcohol once a month. We increased the replacement frequency of the filter membrane and the cleaning frequency of the sampling tube in the event of heavy pollution to ensure that the filter membrane and sampling tube were in a clean state. The length of sampling tube with 6 mm outer diameter was approximately 3 m, the material was PFA with excellent chemical inertness and the

sampling flow rate was 6 SLM meaning that the residence time of the gas in the sampling tube was less than 0.5 s. Besides, the sampling loss was calibrated before the experiment. We assessed the measured spectrum every day to ensure the authenticity of the measurement results. Multiple reflections in the resonator cavity enhanced the length of the effective absorption path, thereby enhancing the detection sensitivity of the instrument. The 1σ detection limits for HONO and NO_2 were about 60 ppt and 100 ppt, respectively, and the time resolution was 1 min. The fitting wavelength range was selected as 359–387 nm. Sample loss and secondary formation of HONO were both considered in this instrument and the measurement error of HONO was estimated to be approximately 9%. The sampling tube was heated to 35℃ and covered by insulation cotton materials to prevent the effect of condensation of the water vapor(Lee et al., 2013).

The uncertainties for the aerosol and NO measurements were10-20%(Tian et al., 2016) and 10%(Xu et al., 2015), respectively. As shown in Figure 1, the NO_2 concentration measured by IBBCEAS had the same trend as the NO_2 concentration measured by TEI 42i, and NO_2 concentration measured by IBBCEAS is always lower than that by TEI 42i. The average NO_2 concentration determined by IBBCEAS and TEI 42i were 14.99 ppb and 18.68 ppb, respectively. Besides, this result also proved by (Villena et al., 2012;Xu et al., 2019;Zheng et al., 2020) that chemiluminescence instruments used for indirect NO_2 detection in monitoring networks were affected by other active nitrogen components. The manuscript has been revised as follows: As expected, the NO_2 concentration measured by IBBCEAS had the same trend as the NO_2 measured by TEI 42i, and NO_2 concentration measured by IBBCEAS was always lower than that by TEI 42i (Figure S1). The reference (Villena et al., 2012) supporting the statement has been added in the manuscript and Figure 1 has been added in the "Supplementary Material" named as Figure S1.

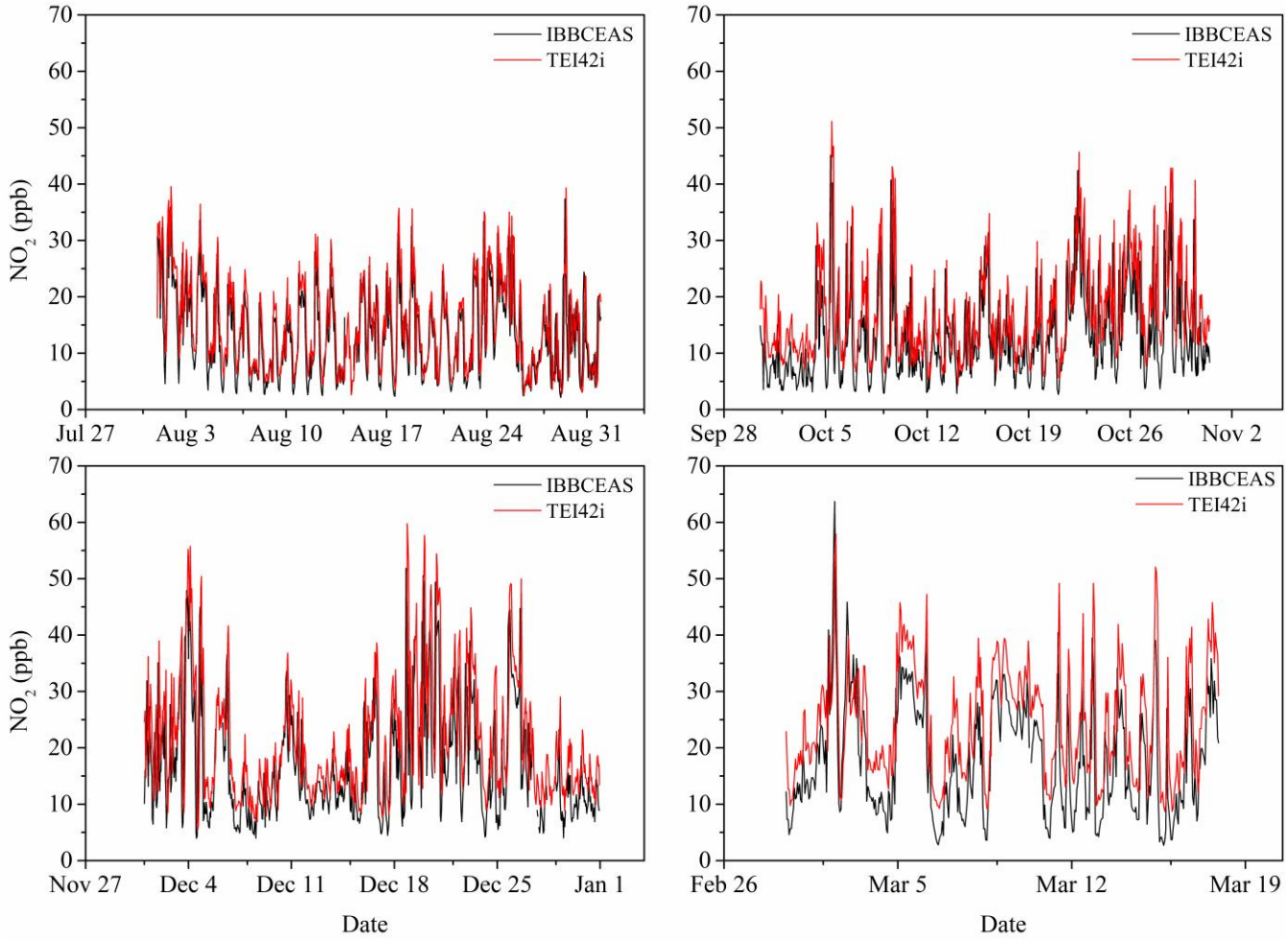

Figure 1. Time series measurements of NO_2 from the IBBCEAS and TEI42i.

The measurement site isn't described adequately. The paper notes that a coastal location, land/sea breeze effects, vehicle exhaust emission, and contributions from diesel traffic are important for understanding HONO abundance, but none of these contributions are detailed here. The conclusion states that site was surrounded by expressways, but these are not detailed in the body of the text. How close are the expressways? Are there diurnal traffic patterns? Figure 1 gives a map, but it does not have sufficient detail to understand the sampling location. The figure should show latitude on an axis, clearly identify land and water, show major roads. And the map should use km rather than miles. The meteorology must be described. Is there a land/sea breeze effect here? What is the mixed layer height? Is the top of the building always within the mixed layer? I expect some nighttime measurements are capturing a residual layer of pollution that may have been processed for longer periods. How large is the city, and what is the proximity of soils and tall buildings (proposed sources of HONO)? The motivation for this paper is that coastal cities have been under sampled, but none of the characteristics important to this coastal location are described.

Response: Thanks for your careful working and constructive comments. The detailed surroundings have been shown in Figure 2 and specific description has been added in Sec. 2.1 (Site description) like this: As shown in Figure.2 (left), Xiamen is located in the southeast coastal area of China and faces the Taiwan Strait in the east. It suffers from sea and land breeze throughout the year with spring and summer more frequently(Xun et al., 2017). The IUE supersite is surrounded by a Xinglin Bay, several universities (or institutes), and several major roads with large traffic fleet, such as Jimei Road, Shenhai Expressway (870 m) and Xiasha Expressway (2300 m) (Figure 2 (right)). Although we did not obtain diurnal traffic patterns in this manuscript, the diurnal variation of NOx can roughly indicate the traffic flow because the main source of NOx in this site is traffic. The figure has shown longitude and latitude on horizontal axis and vertical axis, respectively. Land, water and major roads had been clearly identified. The map had replaced miles with km.

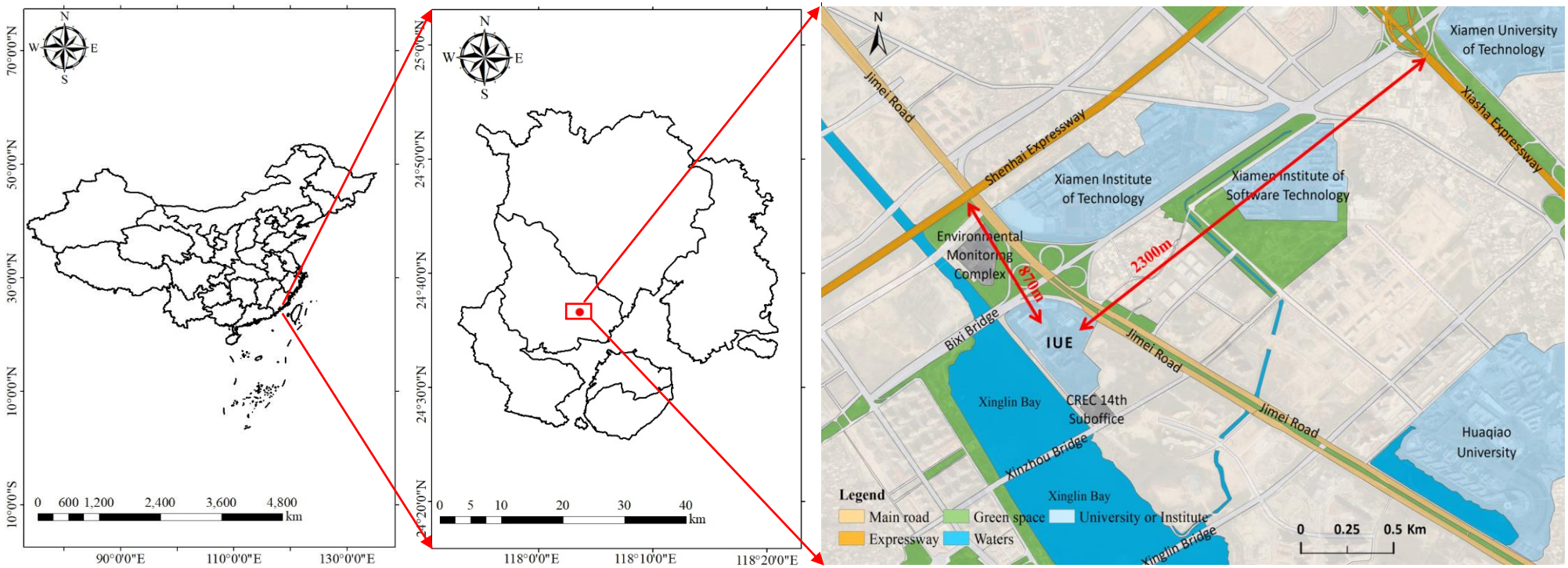

Figure 2. Location of Xiamen in China (left), location of IUE in Xiamen (middle) and surroundings of IUE (right).

The meteorology had been added in the manuscript as follow: Meteorological parameters at 8:00, 11:00, 14:00, 17:00 and 20:00 were applied to estimate atmospheric mixed layer heights by Nozaki method (Cheng et al., 2001).As shown in Figure 3, the average mixed layer heights were 821.59 m, 879.44 m, 1185.28 m and 1059.37 m for spring, summer, autumn and winter, respectively. The minimum values of mixed layer height were 69.30 m, 109.63 m, 282.66 m, and 121.73 m for spring, summer, autumn and winter, respectively, which indicated that the top of the building always within the mixed layer except for spring. As for spring, only one value is lower than the height of sampling site, which further indicates that the top of the building always within the mixed layer during observation period.

The area of Xiamen is 1700.61 km^2 with a population of 4.11 million (http://tjj.xm.gov.cn/tjzl/). The number of motor vehicles in 2018 was 1,572,088, which was 2.73 times as many as ten years ago. The surrounding soil is mainly used for green not for agriculture. The characteristics to this coastal location have been descried in Line 60-65 like this: Field measurements of HONO and its precursor NO_2 at sites with different aerosol load & composition, and relative humidity (RH) are necessary to deepen our knowledge of the HONO formation mechanisms. Such measurements have been carried out in coastal cities in China, including Guangzhou (Qin et al., 2009), Hong Kong (Xu et al., 2015), and Shanghai (Cui et al., 2018), where the air pollution is relatively severe during their research period . However, there has been a lack of research into HONO in coastal cities with good air quality, low concentrations of NO_x and $PM_{2.5}$, but strong sunlight and high humidity. Insufficient research on coastal cities with good air quality has resulted in certain obstacles to assessing the photochemical processes in these areas. Due to different emission-source intensities and ground surfaces, the atmospheric chemistry of HONO in the southeastern coastal area of China is predicted to have different pollution characteristics from those found in other coastal cities.

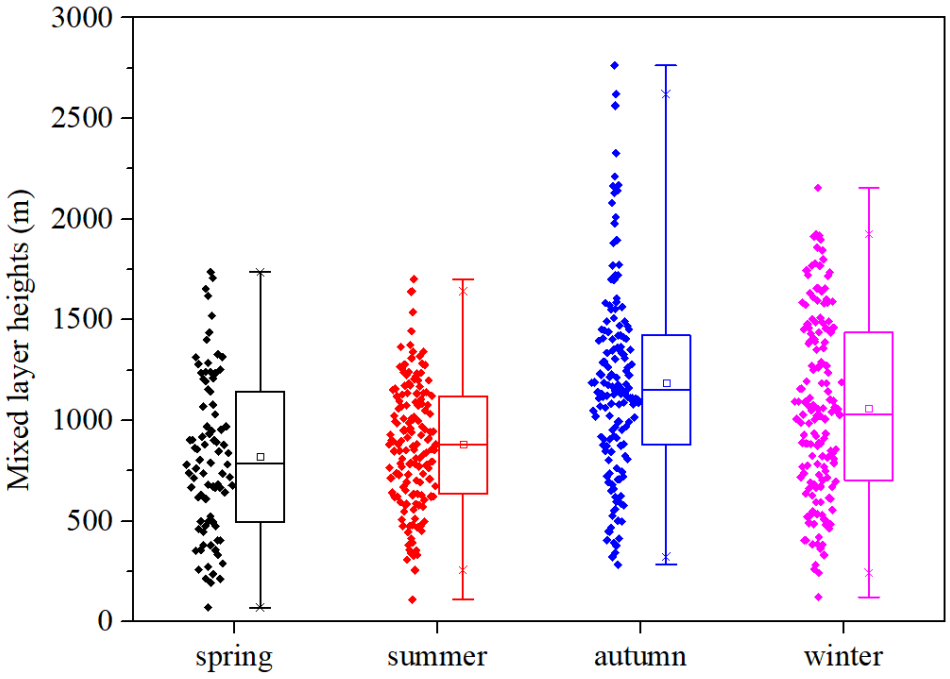

Figure 3. Box plot of mixed layer heights by season. Scattered points represent hourly values of mixed layer heights at 8:00, 11:00, 14:00, 17:00 and 20:00.

Critical ancillary measurements are not adequately reported. What are the Ozone levels at night? The paper reports the average ozone for the entire study, but this doesn't reveal whether ozone is titrated at night, whether there is large ozone production during the day, and the photochemical environment of the measurement location. What is the temperature at this location? Figures 2 and 6 show that the length of day is the same for all seasons,

but this can't be right. On line 99, please describe what you mean with _R and _M in the photolysis rate constants.

Response: Thanks for your valuable suggestion. The critical ancillary measurements, including O_3 and temperature, have been added as follows:

As shown in Figure 4(e), diurnal variations of O_3 concentrations were opposite to that of HONO and NOx, where the minimums were in the morning rush hour because of NO titration(Li et al., 2017), while the maximums were in the afternoon (13:00-15:00 LT) due to high NO_2 photolysis rates and photochemical reactions(Song et al., 2017).The minimum O_3 concentrations in spring, summer, autumn and winter were 13.17, 7.77, 26.99 and 8.33 ppb, respectively. The maximum O_3 concentrations in spring, summer, autumn and winter were 36.46, 60.66, 62.19 and 31.55 ppb, respectively. The O_3concentration in autumn was much higher than that in other seasons, which was explained by the weaker NO titration effect, strong photochemical reaction and region transport. The amplitude was largest in summer with 52.89 ppb, followed by autumn (35.20 ppb), spring (23.29 ppb) and winter (23.22 ppb), which indicated strongest local generation of O_3 in summer, followed by autumn, spring and winter.

As shown in Figure 4(f),The diurnal variations of temperature in the four seasons had the same trend, reaching the maximum at 13:00-14:00 and the minimum at 5:00-6:00. The diurnal maximum average temperatures for spring, summer, autumn and winter were 18.93 ℃, 33.71 ℃, 28.14 ℃ and 22.11 ℃, respectively. The diurnal minimum average temperatures for spring, summer, autumn and winter were 15.24 ℃, 21.12 ℃, 21.28 ℃ and 16.15 ℃, respectively. The average temperature in summer was 30.00 ℃ followed by autumn (24.02 ℃), winter (18.41 ℃), and spring (16.59 ℃).

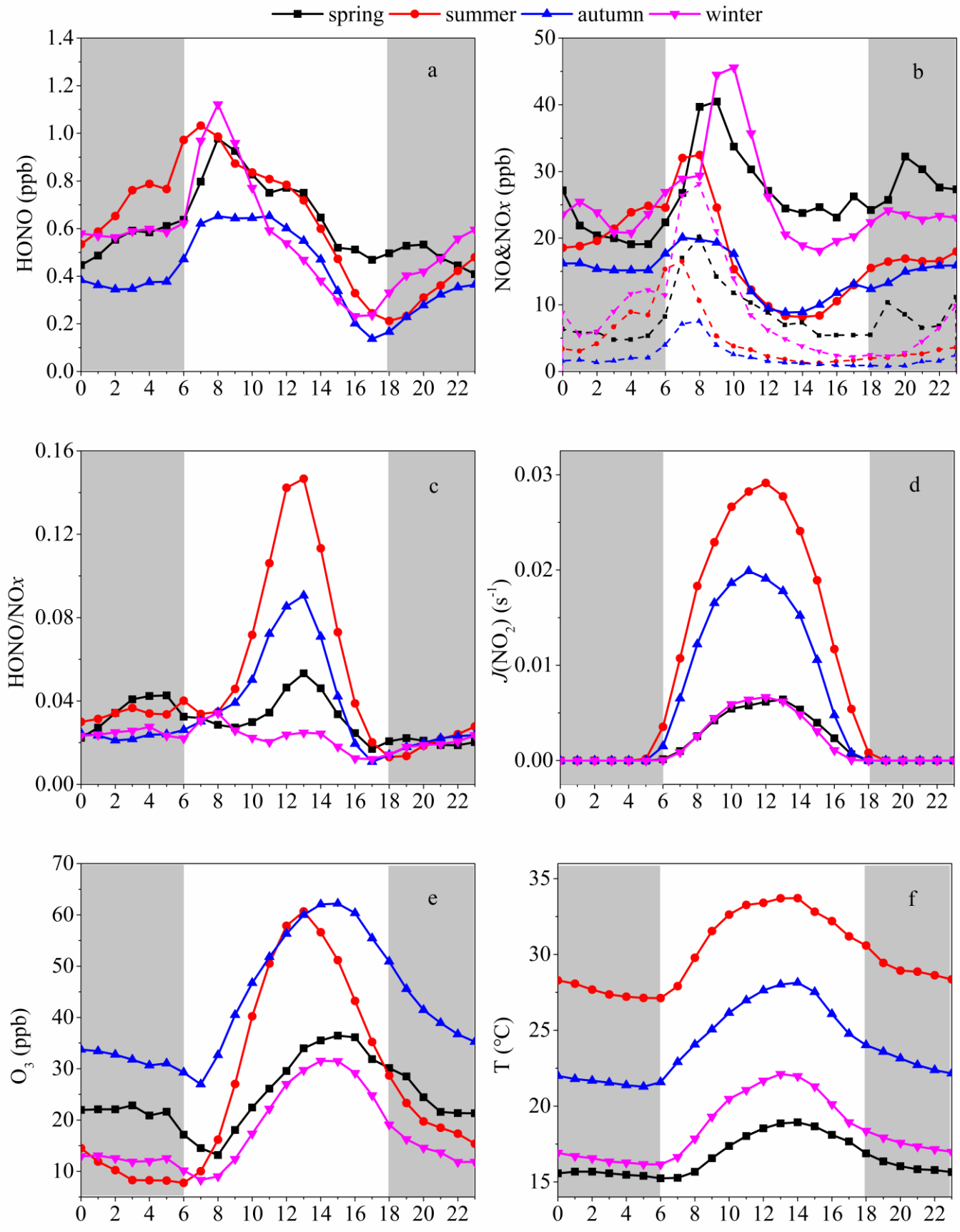

Figure 4. Diurnal variations of (a) HONO, (b) NO (hollow markers and dashed lines) & NOx (solid markers/lines), (c) HONO/NOx, (d) $J(NO_2)$, (e) O_3, and (f) T. The gray shading indicates nighttime (18:00–06:00, including 18:00).

The day lengths of the four seasons were indeed different, which could be inferred by the different lengths of $J(NO_2)> 0$ (Fig.6(d)). In order to be comparable with thosesites located in eastern China with similar longitude(Xu et al., 2015;Li et al., 2018;Liu et al., 2019;Qin et al., 2009),however, the same method was used to distinguish between day and night.The photolysis rate constants with _R and _M represented radical photolysis channel and molecular photolysis channel, respectively. Specifically, HCHO was removed by the reactions (R1) and (R2), and NO_3 was removed by the reactions (R3) and (R4), respectively(Röckmann et al., 2010).

$$HCHO + h\nu \rightarrow CHO + H \qquad J(HCHO\_R) \quad (R1)$$

$$HCHO + h\nu \rightarrow H_2 + CO \qquad J(HCHO\_M) \quad (R2)$$

$$NO_3 + h\nu \rightarrow NO_2 + O^3P \qquad J(NO_3\_R) \quad (R3)$$

$$NO_3 + h\nu \rightarrow NO + O_2 \qquad J(NO_3\_M) \quad (R4)$$

Several of the interpretations are difficult for me to understand and require further analysis. For example, line 264 says “It is hoped that HONO is in the photostationary state. . ..”, and from there, all calculations assume that is the case. The PSS assumption needs to be carefully examined. An analysis of measurements from a similar height on top of a building in Houston show that the PSS assumption may not be correct (Lee et al, Urban measurements of atmospheric nitrous acid: A caveat on the interpretation of the HONO photostationary state, JGR 2013). The Lee et al paper shows that the PSS assumption needs to be carefully examined to quantify the strength of an unknown HONO source. And this is especially true for measurements that are adjacent to major expressways. Table 2 shows fresh vehicle plumes measured during midday with HONO/NOx comparable to nighttime measurements, which may suggest that these plumes are not in PSS. What is HONO/NOx (PSS) for the daytime plumes in Table 2?

Response: Thanks for your comments. The PSS assumption does need to be carefully examined to quantify the strength of an unknown HONO source, but the PSS was not applied to quantify the strength of an unknown HONO source in this manuscript. The result of PSS was a qualitative description of unknown sources. The unknown sources were quantified by budget analysis. A total of 34 cases met these strict criteria for estimation of the HONO vehicle emission ratios, where only one case appeared at midday (11:00-11:15). This phenomenon indicates that majority plumes during midday are not affected by vehicles. Besides, fresh vehicle plumes can be assumed to be of minor importance around noon, as NOx values exhibit a minimum and show low variability(Sörgel et al., 2011).

The HONO production rate from unknown sources reported here is gigantic: 14.78 ppb/h in summer, when it accounted for nearly all HONO production. This number should be compared with previous reports. Have such high values every been reported before? Ryan et al (referenced here) report 1 ppb/hr, and some studies have shown that summer daytime HONO and HONO/NOx can be explained without invoking any unknown source (Lee et al, Urban measurements of atmospheric nitrous acid: A caveat on the interpretation of the HONO photostationary state, JGR 2013; Neuman et al., HONO emission and production determined from airborne measurements over the Southeast U.S., JGR 2016).

Response: Thanks for your comments. The calculated noontime $R_{unknown}$ at Xi'an (0.98 ppb h^{-1}) during summer, and the major loss route of HONO is photodecomposition with an average value of 1.50 ppb h^{-1} around noontime. Average maximum J(HONO) value of 9.5×10^{-4} s^{-1} was obtained at noontime in summer in Xi'an(Huang et al., 2017). The summer noontime $R_{unknown}$ for Beijing was 3.05 ppb h^{-1}, R_{phot} was 4.25 ppb h^{-1}, and $J(NO_2)$ was

$8.0\times10^{-3}s^{-1}$(Wang et al., 2017), while $J(NO_2)$ was $2.91\times10^{-2}s^{-1}$for Xiamen summer noontime. The $J(NO_2)/J(HONO)$ kept relatively constant (5.34~5.69) during daytime in one site, which indicates that J(HONO) of Xiamen was 3.64 times that of Beijing. The summer noontime $R_{unknown}$, $J(NO_2)$, and R_{phot} for Xiamen was 14.78 ppb h^{-1}, $5.34\times10^{-3}s^{-1}$, and 15.76 ppb h^{-1}, respectively. Average maximum J(HONO) value of ~1.5 $\times10^{-3}$ s^{-1} was obtained at noontime in autumn in Xinken, and the $R_{unknown}$ at noontime was ~4.90 ppb h^{-1}, R_{phot} at noontime was ~5.41 ppb h^{-1}(Su et al., 2008). The corresponding autumn noontime values for Xiamen are 3.63 $\times10^{-3}$ s^{-1}, 6.49 ppb h^{-1}, and 7.92ppb h^{-1}, respectively. The major sink is photolysis of HONO, and the major source is unknown. Therefore, unknown source is nearly equal to photolysis of HONO mainly depending on photolysis rate of HONO. The photolysis rate of HONO in summer in Xiamen is significantly higher than other cities. Therefore, it is reasonable that unknown source in summer noontime is gigantic in Xiamen.

Several of the figures are difficult to understand. What are the red lines and dashed lines in figure 8? The logarithmic fits should be described, as they don't appear to encompass the data. It appears that the data could be just as easily fit with a line. What is the color scale on the right? What are the green squares in Figure 10? It would be helpful to keep a consistent color scale for the seasons. All of the figure captions should be expanded to explicitly identify every symbol and line shown on each figure. Labels and units must be included for every axis and colorscale (these are missing on figs 1, 2, 3, 6, 8).

Response: We are sorry to make it difficult to understand and some notes have been added in the manuscript for better understanding as follows: the red lines and dashed lines in figure 8 represents logarithmic fitting curve and turning point, respectively. As shown in Figure 5 and Figure 6, the result of logarithmic fitting was better than that of linear fitting in spring, summer and autumn according to obvious improvement of R^2 in logarithmic fitting. Of course, no matter what kind of fitting, there is no way to make all the observed data fall on the line, only as close as possible to the fitted line. The color scale on the right is the aerosol neutralization degree F. It is a mistake that the green squares in Figure 10, which should be green triangles and has been corrected. Every symbol and line shown on each figure has been explicitly identified in all of the figure captions. Labels and units have been added for axis and colorscale on figs 1, 2, 3, 6, 8.

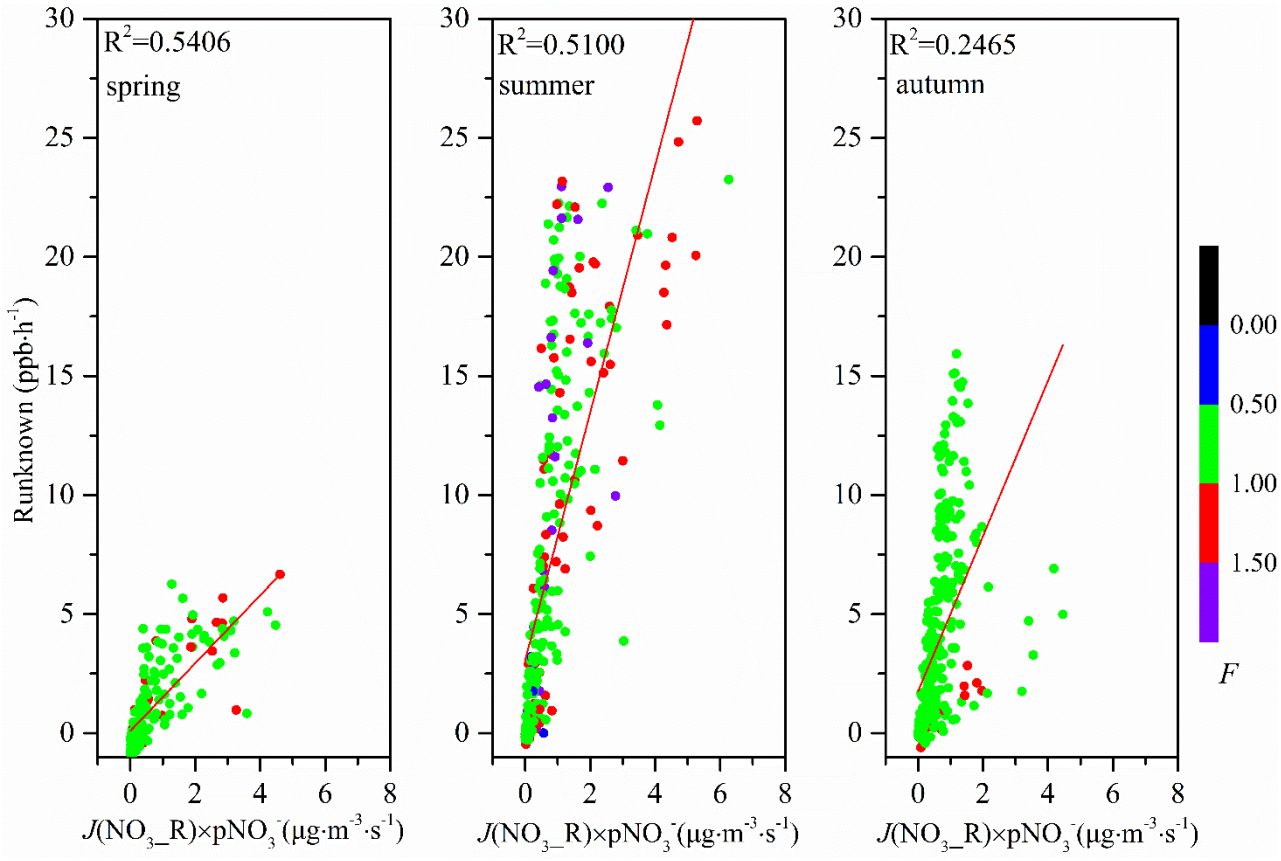

Figure 5. Relationships between the photolysis of particulate nitrate and $R_{unknown}$, colored by F in spring, summer, and autumn. Red lines represent linear fitting curve.

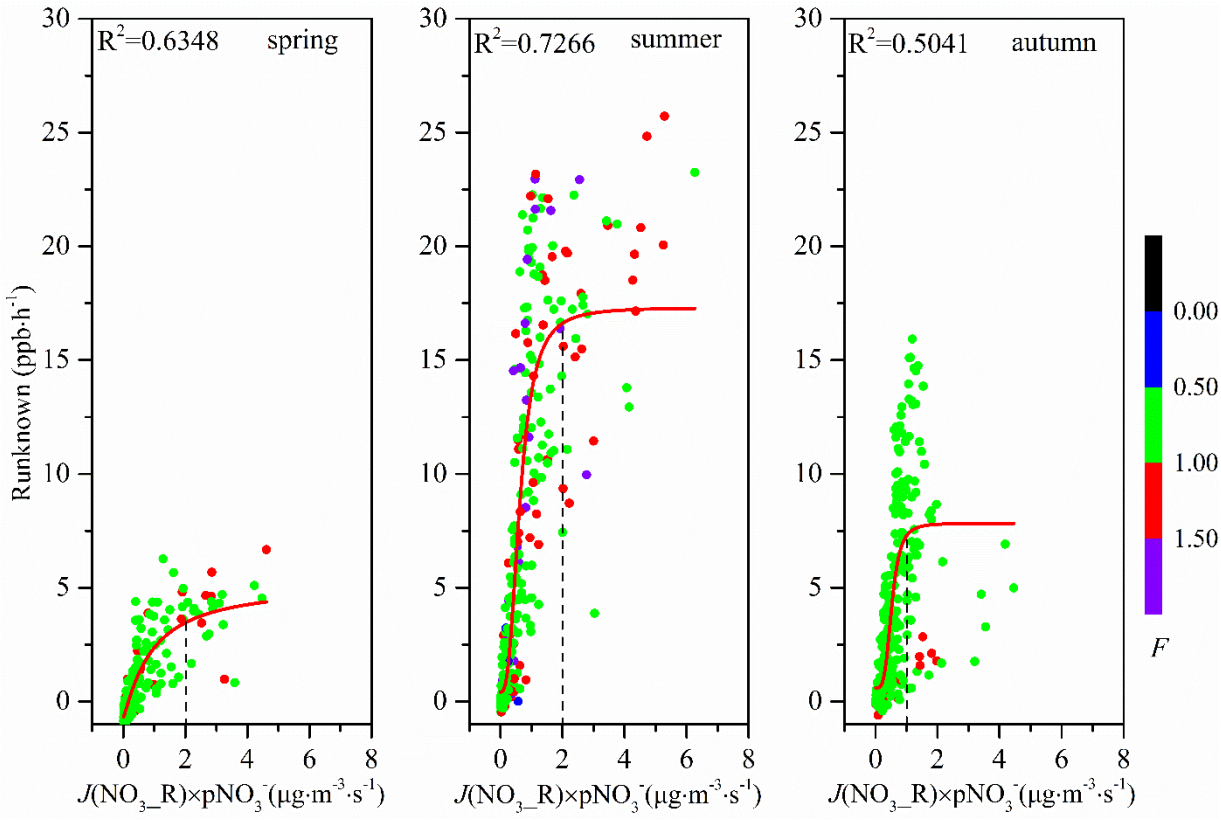

Figure 6: Relationships between the photolysis of particulate nitrate and $R_{unknown}$, colored by F in spring, summer, and autumn. Red lines and dashed lines represent logarithmic fitting curve and turning point, respectively.

I have trouble making sense of the concluding lines of the abstract and conclusion. The conclusion ends (lines 448-450) by stating that HONO provides an OH radical source (4.31 ppb/h) an order of magnitude greater than its concentration (0.66 ppb). I don't understand the comparison of a production rate with a concentration. The order of magnitude increase is also mentioned in the previous section, but I cannot see where this value comes from. The last line of the abstract states the study "draws a full picture of the sources of HONO. . ." But the vast majority of sources are unidentified. A more accurate statement might be that the HONO observations here do not identify the processes that determine HONO chemistry.

Response: Thanks for your valuable suggestion. The statement "HONO provides an OH radical source (4.31 ppb/h) an order of magnitude greater than its concentration (0.66 ppb)." is according to the reference(Ryan et al., 2018). This is not the comparison of a production rate with a concentration but a description about the strong ability of HONO to produce OH radicals. "An order of magnitude" is inaccurate and has been changed to "up to a factor of 5.53"in Line 428 and Line 449. Besides, the statement "The HONO values calculated based on PSS were an order of magnitude small than the observed daytime HONO values" has been changed to "The HONO values observed is 3.86, 9.39, 7.32 and 2.47 times of the HONO values calculated based on PSS for spring, summer, autumn and winter, respectively". The statement "This study draws a full picture of the sources of HONO across all four seasons and improves the comprehension of HONO chemistry in southeastern coastal China." has been changed to "Observation on HONO across four seasons with various auxiliary parameters improves the comprehension of HONO chemistry in southeastern coastal China".

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