Thank you for your careful review and constructive suggestions. These suggestions are quite valuable to us, and help improve our manuscript a lot.

Point-to-point responses

We appreciate the reviewers for their valuable and constructive comments, which are very helpful for the improvement of the manuscript. We have revised the manuscript carefully according to the reviewers' comments. We have addressed the reviewers' comments on a point-to-point basis as below for consideration, where the reviewers' comments are cited in ***black****, and the responses are in* ***blue****.*

A better understanding of the sources and formation mechanisms of HONO is important for understanding troposphere oxidation and processes of secondary pollution. Previous research has focused on the near-surface layer such that there is insufficient literature measuring heterogeneous formation of HONO in the vertical profile. This study uses MAX-DOAS to study the vertical distributions of HONO and its sources over the sea along a Chinese coastline and at coastal stations. Retrievals of vertical profiles of aerosol, NO2 and HONO allow the examination of the differences in heterogeneous production of HONO in sea versus inland cases.
This work can provide important new information on the variation of sources of HONO in the vertical profile within the lower troposphere. However, the evaluation of data uncertainty is incomplete and hinders the use of the study's findings. The detailed comments to be considered are below:

Major Comments

There is general lack of uncertainties or error estimates presented with the measurements throughout, which makes the significance of the findings and conclusions uncertain. Any comparison of averages should include the standard deviations (ex. the VCDs on line 6 and concentrations on line 9). The results and discussion section requires discussion of which results are statistically significant and, therefore, an important contribution to the knowledge of the field. Presenting the uncertainties associated with the retrieved vertical profiles of NO2 and HONO is also required to draw significant conclusions about trends (Lines 269 to 281 & figure 9). The optimal estimation method should have produced some estimate of error when retrieving the vertical profiles. These errors bars should be included in the figures.

Re: Thanks for your great comments.

We have supplemented error analysis in the main text as follows.

Main text:

"2.4 Error analysis

For profile retrieval, the error sources can be divided into four different types: smoothing error, measurement noise error, forward model error, and model parameter error (Rodgers, 2004). However, in terms of this classification, some errors are difficult to be calculated or estimated. For example, forward model error, which is caused by an imperfect representation of the physics of the system, is hard to be quantified due to the difficulty of acquiring an improved forward model. Given calculation convenience and contributing ratios of different errors in total error budget, we mainly took into account error sources based on the following classification, which were smoothing and noise errors, algorithm error, cross section error, and

uncertainty related to the aerosol retrieval (only for trace gas). Here, we estimated the contribution of different error sources to the trace gas vertical column densities (VCDs) and AOD, and near-surface (0–200 m) trace gas concentrations and aerosol extinction coefficients (AECs), respectively. The detailed demonstrations and estimation methods are displayed below, and the final results are summarized in Table 3.

a. Smoothing errors arise from the limited vertical resolution of profile retrieval. Measurement noise errors denote the noise in the spectra (i.e., the fitting error of DOAS fits). They can be quantified by averaging the error of retrieved profiles, as the error of the retrieved state vector equals the sum of these two independent errors. We calculated the sum of smoothing and noise errors on near-surface concentrations and column densities, which were 14 and 5 % for aerosols, 16 and 17 % for NO_2, and 20 and 22 % for HONO, respectively in the sea scene. The corresponding values were 13 and 5 % for aerosols, 14 and 16 % for NO_2, and 18 and 20 % for HONO, respectively at SUST and 13 and 5 % for aerosols, 15 and 17 % for NO_2, and 19 and 21 % for HONO at CAMS.
b. Algorithm error is the discrepancy between the measured and modelled DSCDs. This error contains forward model error from an imperfect approximation of forward function (e.g., spatial inhomogeneities of absorbers and aerosols), forward model parameter error from selection of parameters, and error not related to the forward function parameters, such as detector noise (Rodgers, 2004). Algorithm error is a function of the viewing angle, and it is difficult to assign this error to each altitude of profile. Usually, the algorithm errors on the near-surface values and column densities are estimated by calculating the average relative differences between the measured and modeled DSCDs at the minimum and maximum elevation angle (except 90 °), respectively (Wagner et al., 2004). Considering its trivial role in the total error budget, we estimated these errors on the near-surface values and the column densities at 4 and 8 % for aerosols, 3 and 11 % for NO_2, and 20 and 20 % for HONO, according to Wang et al. (2017) and Wang et al. (2020).
c. Cross section error is the error arising from an uncertainty in the cross section. According to Thalman and Volkamer, (2013), Vandaele et al. (1998), and Stutz et al. (2000), we adopted 4, 3, and 5 % for O_4 (aerosols), NO_2, and HONO, respectively.
d. The trace gas profile retrieval error represents the one, which is sourced from aerosol extinction profile retrieval and propagated to retrieved trace gas profile. This error could be roughly estimated based on a linear propagation of the total error budgets of the aerosol retrievals. The errors of trace gases were roughly estimated at 15% for VCDs and 10% for near-surface concentrations for the two trace gases in the sea scene. The corresponding values were 14 and 10 % for near-surface concentrations and VCDs, respectively at SUST, and 14 and 10 % at CAMS.

The total uncertainty was calculated by adding all the error terms in the Gaussian error propagation, and the final results were listed in the bottom row of Table 3. We found that the sum of smoothing and noise errors played a dominant role in the total uncertainty.”

Table 3. Error budget estimation (in %) of the retrieved near-surface (0–200 m) trace gas concentrations and AECs, and trace gas VCDs and AOD.

			Error source				Total
			Smoothing and noise errors	Algorithm error	Cross section error	Related to the aerosol retrieval (only for trace gases)	
Cruise route	Near-surface	aerosol	14	4	4	-	15
		NO_2	16	3	3	15	22
		HONO	20	20	5	15	32
	VCD or AOD	AOD	5	8	4	-	10
		NO_2	17	11	3	10	23
		HONO	22	20	5	10	32
SUST	Near-surface	aerosol	13	4	4	-	14
		NO_2	14	3	3	14	20
		HONO	18	20	5	14	31
	VCD or AOD	AOD	5	8	4	-	10
		NO_2	16	11	3	10	22
		HONO	20	20	5	10	30
CAMS	Near-surface	aerosol	13	4	4	-	14
		NO_2	15	3	3	14	21
		HONO	19	20	5	14	31
	VCD or AOD	AOD	5	8	4	-	10
		NO_2	17	11	3	10	23
		HONO	21	20	5	10	31

We have added error bars in all the retrieval vertical profiles (i.e., Fig. 9-11) as follows.

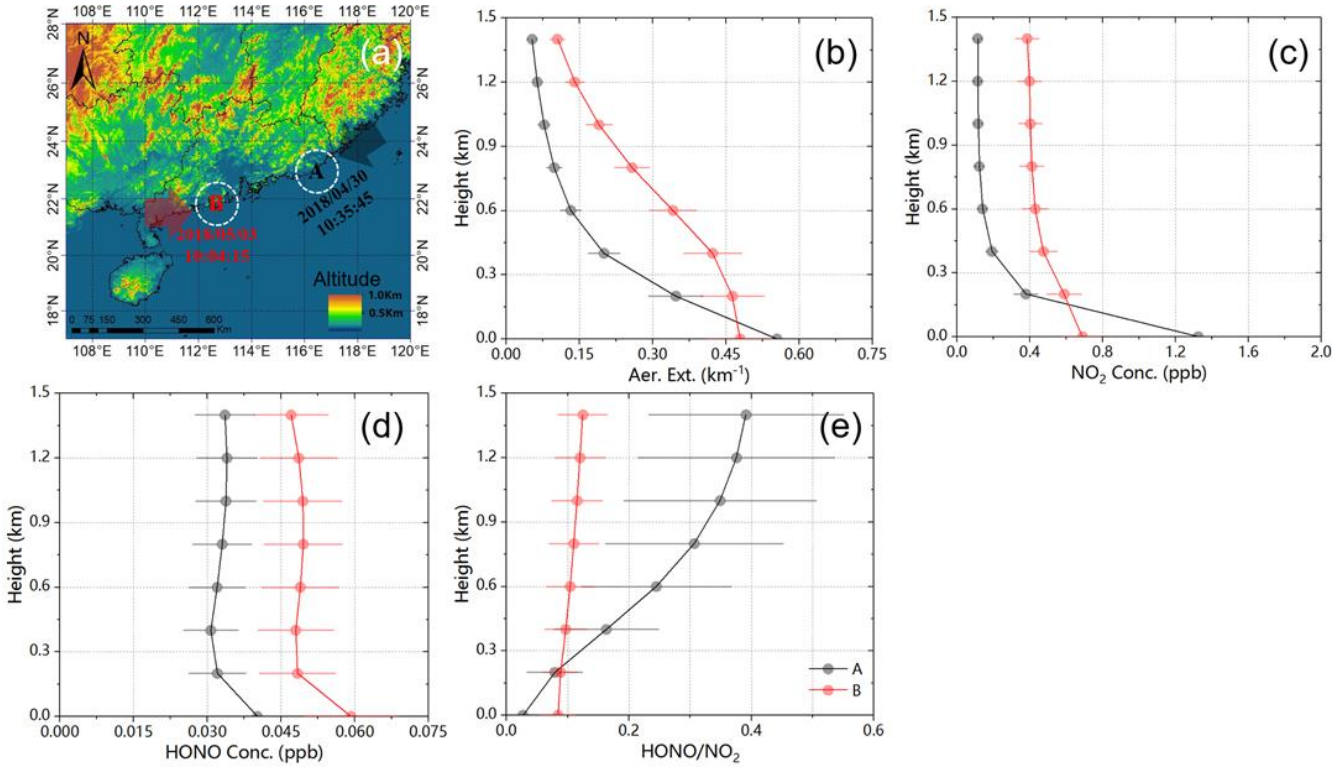


Figure 9. Map (a) shows the two measurement points (A: black, sea-oriented with sea wind; B: red, land-oriented with land wind) during the campaign. Plots (b)–(e) show the vertical profiles of aerosol, NO2, HONO, and HONO/NO2 ratios in the above two measurement points, respectively.

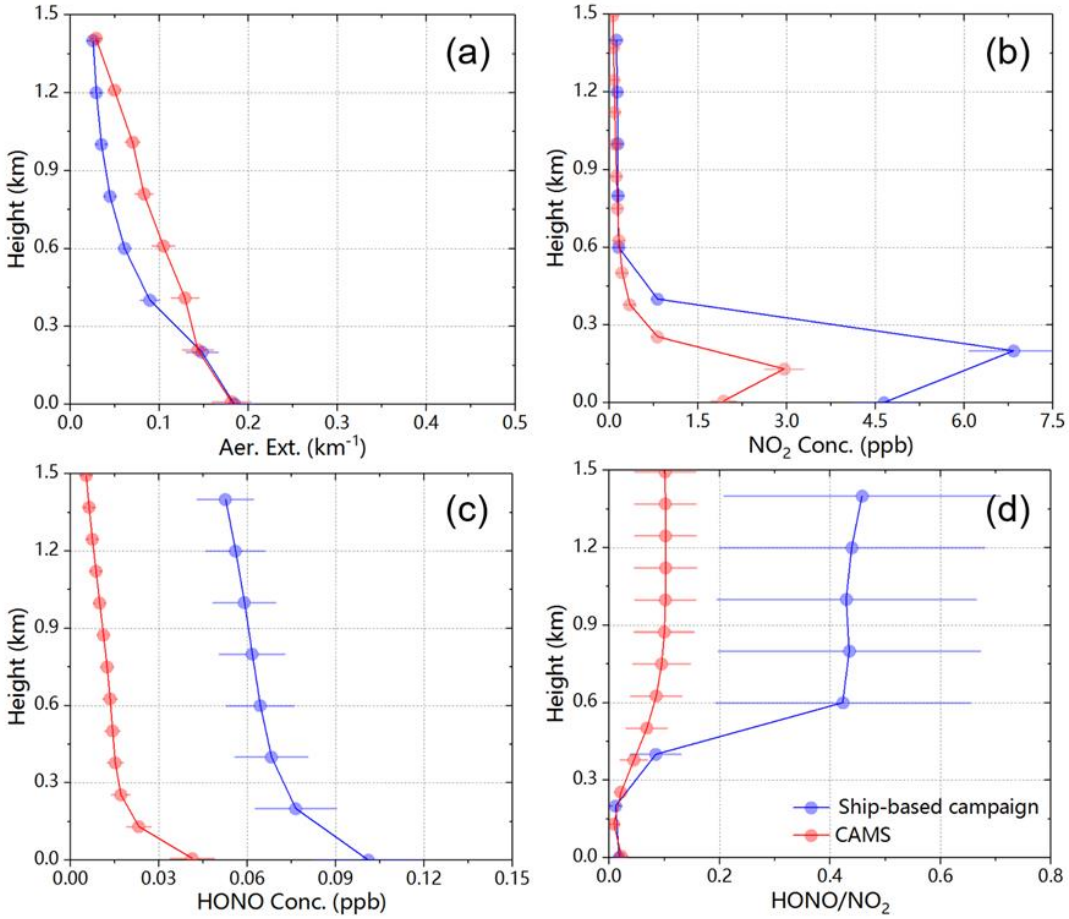


Figure 10. Plots showing the vertical distributions of (a) aerosol extinction, (b) NO2 concentration, (c) HONO concentration, and (d) HONO/NO2 ratio. The blue and red lines represent a ship-based campaign case and a CAMS case, respectively.

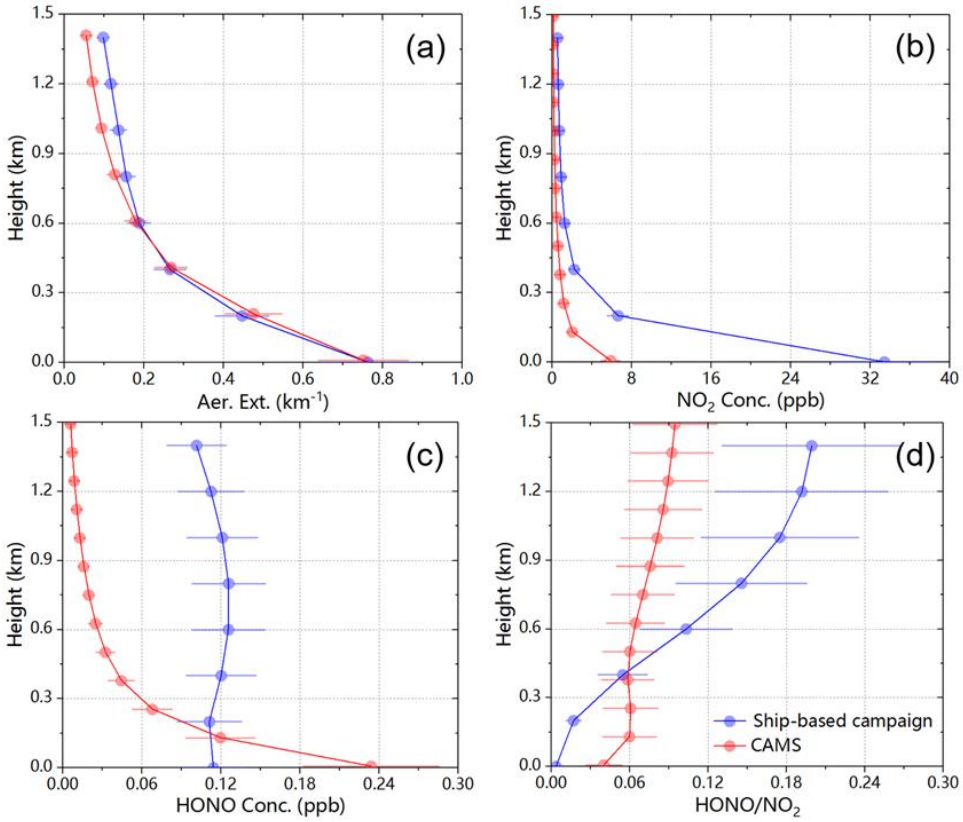


Figure 11. Plots showing the vertical distributions of (a) aerosol extinction, (b) NO2 concentration, (c) HONO concentration, and (d) HONO/NO2 ratio. The blue and red lines represent a ship-based campaign case and a CAMS case, respectively.

Retrieval uncertainty is particularly important for the HONO/NO2 ratios due to error propagation. For example, since the sensitivity of the MAX-DOAS retrievals tend to decrease with increasing altitude, the ratio values at higher altitudes in the profile may be a function of the chosen a-priori values rather than the true state of the atmosphere, and therefore cannot be interpreted. In general, a discussion should be included on how the changing MAX-DOAS sensitivity with altitude impacts the shape and magnitude of the retrievals compared to the true atmosphere (either the methodology or results sections). Otherwise, the readers might assume that the MAX-DOAS vertical profiles are more accurate at higher elevations than is the case (versus, for example, the accuracy level of lidar vertical profiles of aerosol extinction). An example of a typical averaging kernel from the optimal estimation retrieval should thus be provided (ex. in the supplemental). Finally, what sensitivity testing was conducted in terms of the effect on the chosen a-priori shape on the shape of the retrieved profiles?

Re: Thanks for your great comments. We have added corresponding contents in main text and Supplementary materials as follows.

"In this study, an exponential decreasing a priori with a scale height of 1.0 km was used as the initial profile for both the aerosol and trace gases retrieval (Figure S4). The surface concentrations of aerosol, NO_2, and HONO were set to 0.2 km^{-1}, 3.0 ppb, and 1.0 ppb, respectively. We assume a fix set of aerosol optical properties with asymmetry parameter of 0.69, a single scattering albedo of 0.90, and ground albedo of 0.05. Furthermore, the uncertainty of the aerosol and trace gases a priori profile was set to 100% and the correlation length was set to 0.5 km. The averaging kernels indicated that the sensitivity of the profile retrieval tended to decrease with increasing

altitude, and was especially sensitive to the layers within 0–1.5 km (Figure S5). The sum of the diagonal elements in the averaging kernel matrix is the degrees of freedom (DOF), which denotes the number of independent pieces of information contained in the measurements."

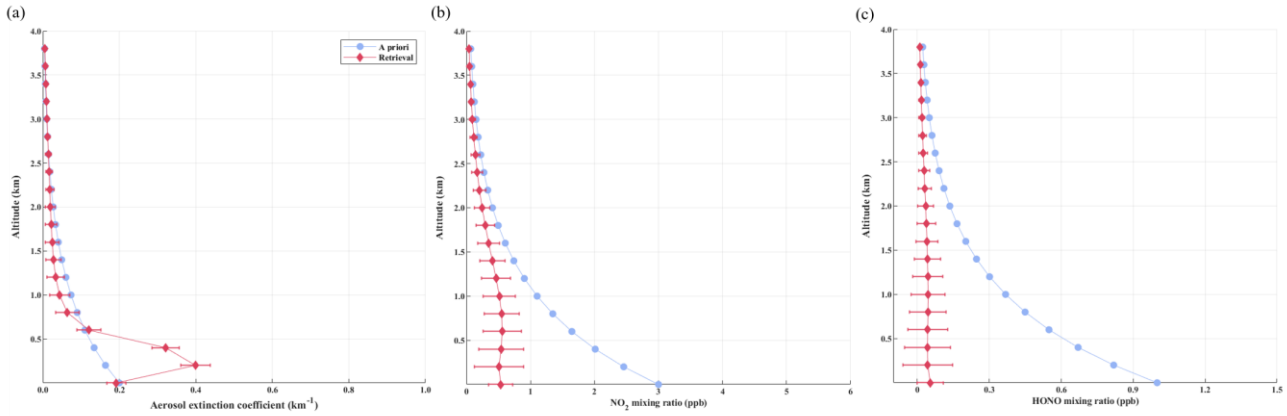


Figure S4. An example of the a priori and retrieved profiles from MAX-DOAS measurements in ship-based campaign (May 1, 2019 at 08:02 LT) for (a) aerosol extinction, (b) NO_2, and (c) HONO.

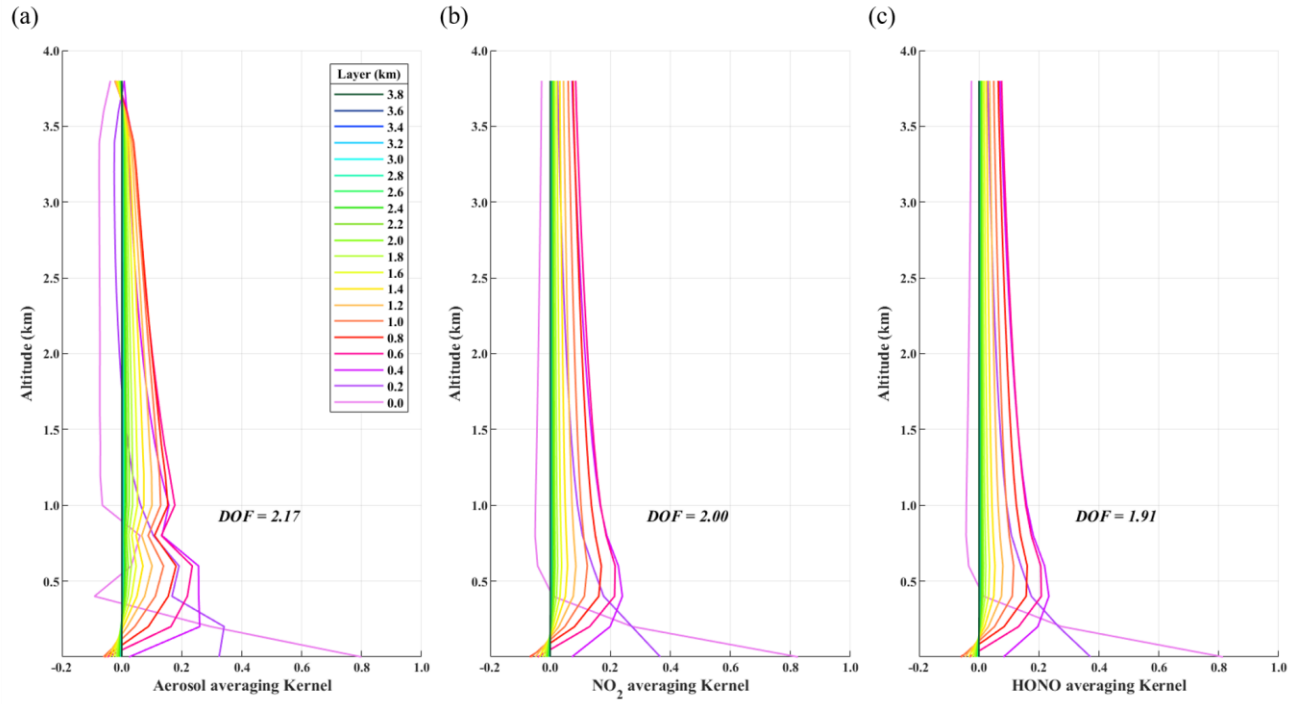
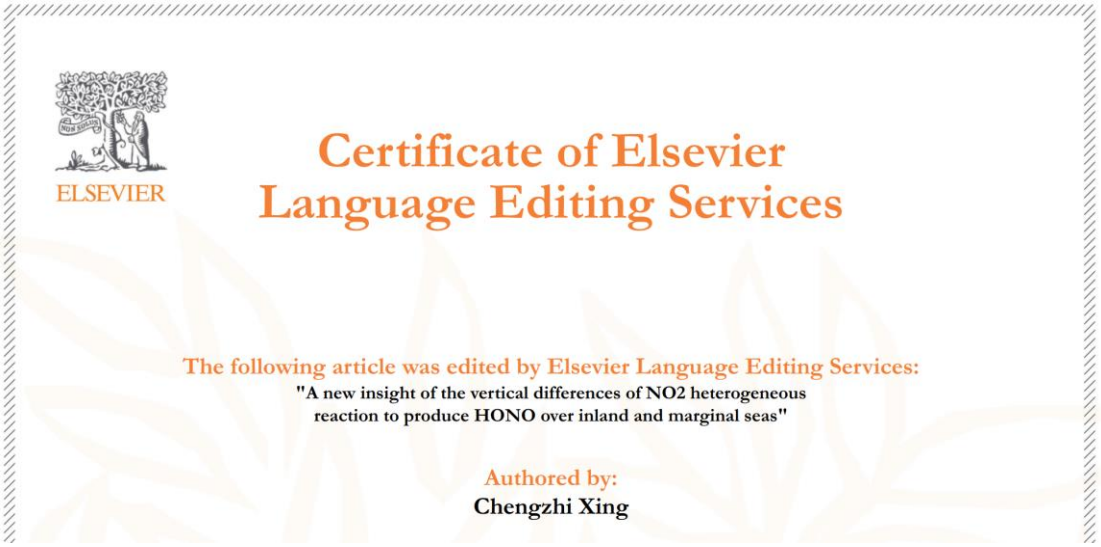


Figure S5. An example of averaging kernel results from MAX-DOAS measurements in ship-based campaign (May 1, 2019 at 08:02 LT) for (a) aerosol extinction, (b) NO_2, and (c) HONO.

The chosen a-priori profile shape really affects the shape of the retrieved profiles. In previous studies, we compared the results retrieved using four different a priori profiles (i.e., linearly, exponential, Boltzmann, and Gaussian profiles) and selected the most suitable one——Gaussian a priori profile (Xing et al., 2017). The aerosol extinction profile retrieved using the Gaussian a priori profile shows the best agreement with simultaneous lidar and balloon-based measurements.

We did a sensitivity test to estimate the uncertainty related to the choice of the a priori profile for retrieved results by varying the scaling height (either 0.5 km or 1 km) (Hendrick et al., 2014). We found that the relative change was within 30%.
Given the effects of the a priori profile, current studies mostly set a priori profiles based on the data from other sources (e.g., modelling and balloon-based in situ measurements). Therefore, it is significant to study and control the impacts of the a priori profile on the retrieved profiles. We plan to do improvements on this aspect in the future. The exact methods include estimating the surface concentrations and VCDs from the DSCDs at the lowest elevation angle and the highest elevation angle, respectively. This method can reduce the dependence of retrieval on other data and enhance the robustness of retrieval procedure.

2. The use of English needs some improvement. Typos and grammatical errors, such as missing the words “that” and “the” in many sentences, reduces overall clarity. The manuscript would benefit from professional English editing.
Re: Thanks for your important comments. We have let a language revision institution help us polish our language and correct mistakes.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Language Editing Services

The following article was edited by Elsevier Language Editing Services:
"A new insight of the vertical differences of NO2 heterogeneous reaction to produce HONO over inland and marginal seas"

Authored by:
Chengzhi Xing

Minor Comments
1. Lines 16 to 19. For improved English, these sentences should use the format “the HONO/NO2 ratio was observed to decrease with increasing temperature…”
Re: Thanks for your great comments.
“The $HONO/NO_2$ decrease along with the increase of temperature, and with peak values on ~12.5℃ in CAMS. The $HONO/NO_2$ increased along with increasing temperature, and with peak values on ~31.5℃ in SUST. In sea case, the $HONO/NO_2$ increased along with the increase of temperature with a peak value on ~25.0℃ under the temperature being larger than 18.0℃.” -> “As temperature increased, the $HONO/NO_2$ ratio decreased with peak values appearing at ~12.5℃ in CAMS, whereas the $HONO/NO_2$ gradually increased and reached peak values at ~31.5℃ in SUST. In the sea case, when the temperature exceeded 18.0℃, the $HONO/NO_2$ ratio rose with increasing temperature and achieved its peak at ~25.0℃.”

2. Lines 29-30. Under land or sea conditions?
Re: Thanks for your great comments. They are all under land conditions. If we don’t add any special instructions, land conditions are the default.

3. Lines 34-35. Amend to "… nitrate amines that pose a threat to human health".
Re: Thanks for your great comments.
"In addition, as a nitrosating agent, HONO can produce carcinogenic nitrite amines to threat to human health (Zhang et al., 2015)." -> "Additionally, as a nitrosating agent, HONO can produce carcinogenic nitrite amines that pose a threat to human health (Zhang et al., 2015)."

4. Line 37. Suggest listing the important/known HONO formation reactions, similar to the introduction in the Wen et al. (2019) referenced.
Re: Thanks for your great comments. We have revised and changed our descriptions as follows.
"Photolysis of HONO in near ultraviolet bands (Eq. 1) is a substantial source of hydroxyl radicals (OH radicals), which are one of the most important oxidants in the tropospheric atmosphere."
"Currently, the known sources of HONO mainly include direct emissions from vehicles, ships, biomass burning and soil, the homogeneous reaction of NO and OH radicals (Eq. 2), the nighttime and daytime heterogeneous reaction of NO_2 (Eq. 3) on aerosols, vegetation, ground and other types of surfaces, and the photolysis of nitrate particles (Eq. 4) (Alicke et al., 2003; Stemmer et al., 2006; Indarto et al., 2012; Wang et al., 2015; Salgado and Rossi, 2002; Zhou et al., 2011)."

$$HONO + h\nu \rightarrow NO + \cdot OH\ (\lambda < 400nm) \quad (1)$$

$$\cdot OH + NO + M \rightarrow HONO + M \quad (2)$$

$$2NO_2 + H_2O \rightarrow HONO + HNO_3 \quad (3)$$

$$HNO_3 / NO_3^- + h\nu \rightarrow HONO / NO_2^- + O \cdot (\lambda \sim 300nm) \quad (4)$$

5. Lines 39-40. Should this be "there are sources of HONO that are poorly understood"? Rewrite for clarity.
Re: Thanks for your great comments.
"There are also some obvious unknown HONO sources (Fu et al., 2019)." -> "Sources of HONO exist that are poorly understood (Fu et al., 2019)."

6. Lines 68 – 69. Are "favourable weather conditions" sea breeze conditions? Otherwise, what does "favourable" mean here?
Re: Thanks for your great comments.
"The formed HONO is completely likely to be transported to land cities at night under favorable weather conditions." -> "The formed HONO is likely to be carried to land cities at night by sea breeze, which will affect the atmospheric oxidation and air quality, and even endanger human health."

7. Line 82. Add "above surface" after 120 m.
Re: Thanks for your great comments. We have added some cited literatures and changed descriptions.
"…and found the maximum value of HONO appeared at 120 m sourced from the heterogeneous reaction of NO_2 on aerosol surface under haze conditions." -> "Taking tower and aircraft as platforms, these techniques were performed to measure HONO vertical profiles, and it was found that the peak values of HONO usually appeared under 200 m at urban and suburban areas (Kleffmann et al., 2003; Stemmler et al.,

2006; Zhang et al., 2009; Wong et al., 2012; Meng et al., 2020; Zhang et al., 2020). These studies also revealed that the heterogeneous reaction of NO_2 on multiple surfaces (ground and aerosol etc.) was an important source of HONO under planetary boundary layer (PBL), especially in haze days. Furthermore, they also reported that the HONO/NO_2 ratios usually decreased with the increase of height under 200 m at inland and coastal areas."

8. Lines 145 – 152. Suggest adding more detail about the optimal estimation method. For example, that the aerosol vertical profiles are retrieved from the O4 DSCDs, which are then used as model inputs for retrieving trace gas vertical profiles. What was the magnitude of the a priori for the aerosol and trace gas retrievals? How many minutes of measurements were included in the retrieval of one profile?

Re: Thanks for your great comments. We have supplemented some contents in Section 2.3 and Supplementary materials as follows.

Section 2.3: "The detailed retrieval procedure is displayed in Appendix I and Figure S3."

Appendix I: "The maximum a posteriori state vector $\mathbf{x}$ is determined by minimizing the following cost function χ^2.

$$\chi^2 = (\mathbf{y} - F(\mathbf{x},\mathbf{b}))^T \mathbf{S}_\varepsilon^{-1} (\mathbf{y} - F(\mathbf{x},\mathbf{b})) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) \quad (1)$$

Here, $F(\mathbf{x},\mathbf{b})$ is the forward model, which describes the measured DSCDs $\mathbf{y}$ as a function of the retrieval state vector $\mathbf{x}$ (i.e., aerosol and trace gas vertical profiles) and the meteorological parameters $\mathbf{b}$ (e.g., atmospheric pressure and temperature profiles); $\mathbf{x}_a$ denotes the a priori vector that serves as an additional constraint; $\mathbf{S}_\varepsilon$ and $\mathbf{S}_a$ are the covariance matrices of $\mathbf{y}$ and $\mathbf{x}_a$, respectively. The retrieval of vertical profiles of aerosols and trace gases were classified into two steps (Figure S3). First, we retrieved vertical aerosol profiles based on a series of retrieved O4 DSCDs at different elevation angles. Second, the retrieved aerosol extinction profiles were utilized as the input parameters to the RTM to retrieve NO2 and HONO vertical profiles. Each scanning sequence of DSCD results (~5.5 min) correspond to one retrieved vertical profile information. In this study, we separated the atmosphere into 19 layers from 0 to 3.8 km with a vertical resolution of 0.2 km. Given the low sensitivity of MAX-DOAS measurements to high altitude and low concentration of pollutants above 3.0 km, we only displayed the vertical profiles below 3.0 km in this work."

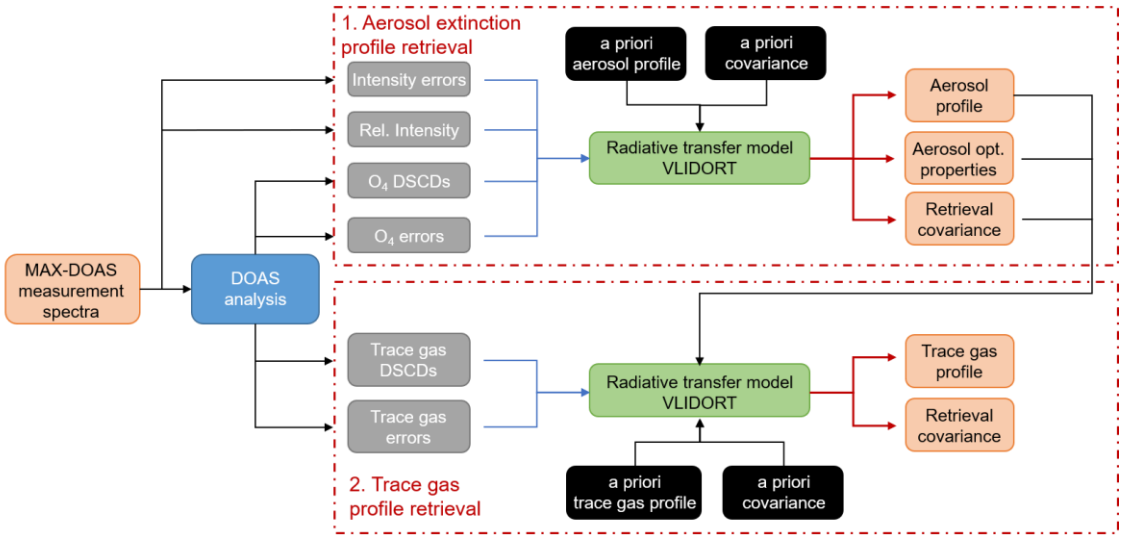


Figure S3. Flowchart of the aerosol and trace gas retrieval algorithm. The dashed-lined red boxes denote the retrieval steps: aerosol and trace gas profile retrieval.

9. Lines 160 – 161. It says in section 3.1 that the radiative transfer model SCIATRAN was used to convert SCDs of NO2 and HONO to VCDs, but why were the vertical profiles retrieved using the optimal estimation method not used to calculate VCDs? This appears to be duplication of work. If there is a lack of confidence in the vertical profiles from the optimal estimation method, this should be explained. Were the VCDs calculated using the two different methods compared? If so, please include in the supplemental and justify the methodological choice.

Re: Thanks for your great comments. The SCDs was converted to VCDs using Eq. c1.

$$VCD = \frac{SCD}{AMF} \quad \text{(c1)}$$

Here, AMF can be simulated by radiative transfer model SCIATRAN, and then we got VCDs of each trace gas. We didn't calculate VCDs using profiles retrieved for two reasons below.

On one hand, one profile is retrieved from one scanning sequence of DSCDs, which contains 11 DSCDs result at different angles (i.e., 1º, 2º, 3º, 4º, 5º, 6º, 8º, 10º, 15º, 30º and 90º). In other words, we could only get one VCD from 11 SCDs if we calculate VCDs using retrieved profile. Comparatively, using simulated AMF to calculate VCDs can let us get one VCD from each SCD result, and we can get more data points along this cruise line. On the other hand, calculating VCDs from one profile means that we need to use concentration at each height to represent one layer's average concentration (Eq. c2), which would introduce larger uncertainties. And there are uncertainties in profile retrieval as well. Therefore, this indirect converting method will bring in many uncertainties, which largely affects the VCD results accuracy.

$$VCD = \sum_{i=0}^{20} (C_i \times H_i) \quad \text{(c2)}$$

We didn't calculate VCDs using the two different methods compared. Instead, we just use Eq. c1 to calculate VCDs for more data points and better data quality.

10. Line 172. Suggest changing "elevated" to "enhanced" to make it clear that the hotspots were much greater than background as opposed to elevated above the surface (i.e. "lifted").

Re: Thanks for your great comments.

"five elevated tropospheric NO_2 VCDs hot spots" -> "Five enhanced tropospheric NO_2 VCDs hot spots were observed…"

11. Lines 171 to 182. A bar chart comparing the averaged NO2 and HONO (with standard deviations) in the five areas will be useful for the reader in terms of observing differences in the distributions described in the text. Box and whisker plots might also be a good choice since they provide more details about outliers.

Re: Thanks for your great comments. We have added corresponding box plots in Supplementary materials as follows.

Main text: "The averaged NO_2 VCDs in above five areas reached 1.07×10^{16}, 1.30×10^{16}, 7.27×10^{15}, 5.34×10^{15}, and 3.12×10^{15} molec. cm^{-2}, respectively (Figure S6(a)). HONO exhibited similar spatial distribution characteristics as NO_2, and the averaged HONO VCDs in above five hot-spot areas reached 1.01×10^{15}, 7.91×10^{14}, 6.02×10^{14}, 5.36×10^{14}, and 5.17×10^{14} molec. cm^{-2}, respectively (Figure S6(b))."

Supplementary materials:

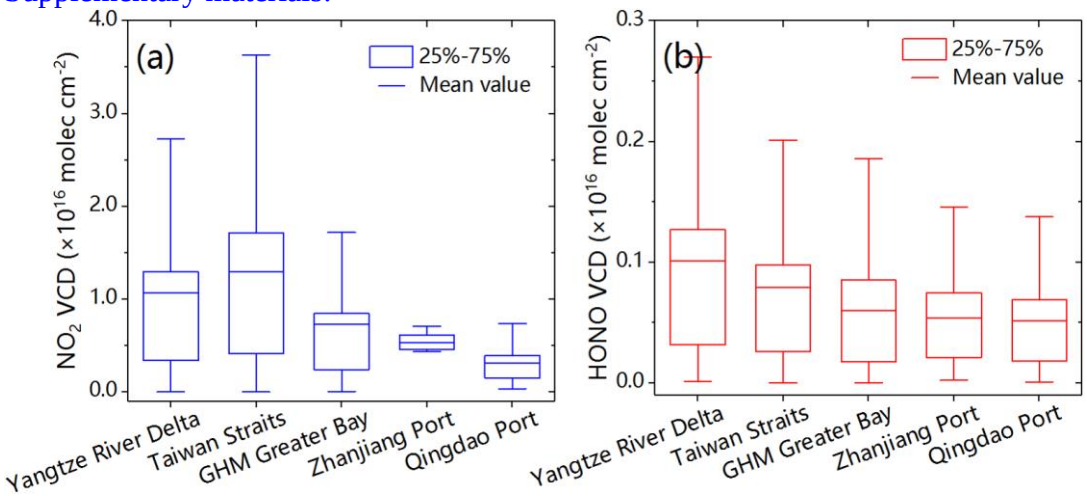


Figure S6. The VCD distribution of (a) NO_2 and (b) HONO for five high-level emission sources (i.e., the coastal areas of Yangtze River Delta, Taiwan straits, Guangzhou-Hong Kong-Macao Greater Bay areas, Zhanjiang Port, and Qingdao port) along the cruise.

12. Line 213. What is meant by "navigation areas"?

Re: Thanks for your great comments. The "navigation areas" meant the shipping routes and international ports. To avoid misunderstanding, we have changed this sentence as follows.

"Sun et al. (2020) reported that HONO concentrations could increase up to 40–100% over the navigation areas" -> "Sun et al. (2020) reported that HONO concentrations could increase up to 40–100% over the shipping routes and international ports…"

13. Lines 210 to 11. More details about these stations are required. Where they located (ex. latitude and longitude coordinates). What are the characteristics of the stations? For example, local pollution sources, topography, prevailing meteorological conditions, etc.

Re: Thanks for your great comments. We have added station information in Section 2.1 the measurement cruise and Supplementary materials as follows. The topography and distribution of two stations are obviously revealed in Figure S2.

Section 2.1: "To fully understand the differences of the impacts of RH, temperature, and aerosol on the HONO secondary formation in land and sea conditions, the Chinese Academy of Meteorological Sciences (CAMS) and Southern University of Science and Technology (SUST) MAX-DOAS stations were selected as inland and coastal areas for analysis, respectively. CAMS is located in the urban of Beijing (116.32°E, 39.94°N), and SUST is located in Shenzhen (114.00°E, 22.60°N) (Figure S2)."

Supplementary materials:

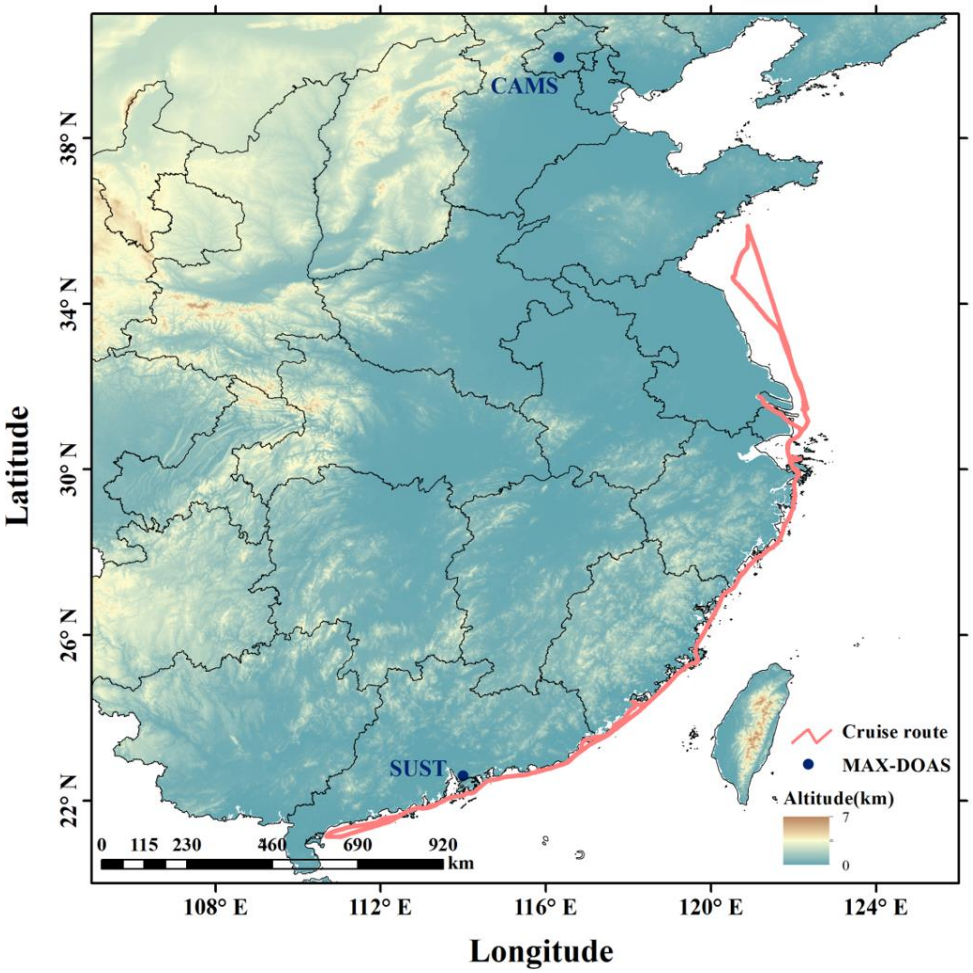


Figure S2. Cruise route and the location of two MAX-DOAS stations (CAMS and SUST).

14. Lines 216 to 222. Much more detail is needed in terms of how the emission ratios were determined (i.e. in the methodology section). Simply citing the literature is not sufficient.

Re: Thanks for your great comments. We have added some details and revised some descriptions as follows.

Main text: "By subtracting the average marine background of NO_x and HONO from the ship plume emission values, the impact of background values is reduced and the emission ratio of ΔHONO/ΔNOx can be obtained, and this emission ratio can be used for quantifying the primary HONO (Sun et al., 2020; Xu et al., 2015). In this study, we used an averaged 0.46±0.31% emission ratio of ΔHONO/ΔNOx referring to Sun et

al. (2020) to understand the primary source of HONO on the sea surface during the campaign."

"Additionally, the calculation method of emission ratios of ΔHONO/ΔNOx in CAMS and SUST was referred from Xu et al. (2015), Liu et al. (2018), and Xing et al. (2021) (Appendix II)."

Supplementary materials:

"Appendix II: The criteria of the identification of fresh plumes

The fresh plumes were selected using the following criteria: (a) [NO_x]>40 ppb, (b) NO/NO_x>0.85, (c) good correlation performing between HONO and NO_x (R>0.90), (d) short duration of plumes (<=2.0 h), and (e) 70^o<SZA<75^o.

MAX-DOAS performed based on the collected solar scattering spectrum to retrieve aerosol, NO_2 and HONO. In general, we believed that the retrieved MAX-DOAS data was reliable, when SZA was not large than 75^o. In order to reduce the influence of fast photolysis of HONO and NO_2, we usually selected data with 70^o<SZA<75^o to calculated $HONO/NO_x$ ratios from direct emission. In this condition, the photolysis rate of NO_2 was not large than 0.25×10^{-3} s^{-1}."

15. Line 223. Since later sections show how meteorological variables impact the HONO/NO2 relationship, it would be helpful to add visualise the impact on these scatterplots using coloured marker points. For example, you could provide versions of these plots where each point has a color corresponding to the temperature. These plots may help to explain some of the outliers that are reducing the R values.

Re: Thanks for your great comments. We have added corresponding Figure in Supplementary materials and demonstrations in main text as follows.

Main text: "The corresponding temperature and RH conditions of each spot are displayed in Figure S8, which roughly reveals the impact of RH and temperature on the process of NO_2 forming HONO through heterogeneous reactions."

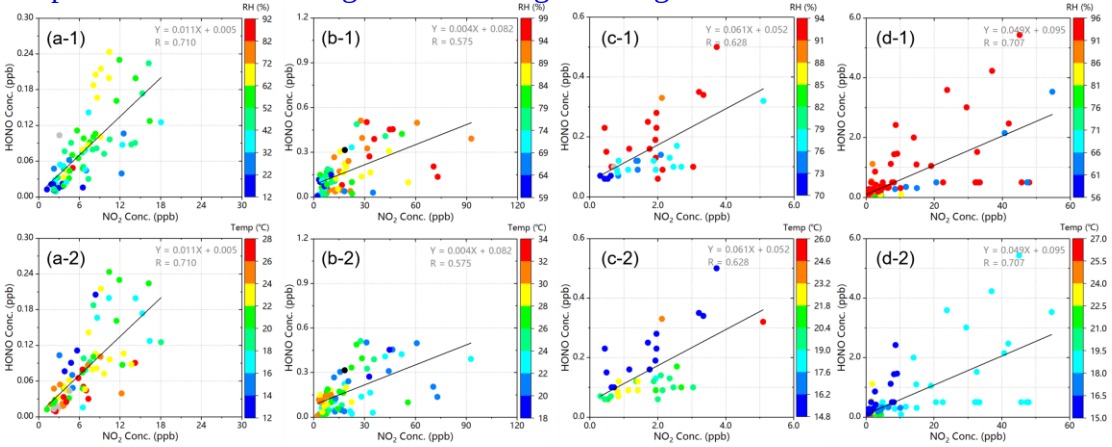


Figure S8. Scatter plots of HONO concentration vs. NO_2 concentration coloured by (1) relative humidity (RH) and (2) temperature in (a) CAMS, (b) SUST, and ship-based measurements of (c) sea-oriented and (d) land-oriented under static weather condition.

However, the meteorological data of CAMS and SUST stations only have the RH and temperature data of 08:00, 11:00 and 14:00. Thus, there are different numbers of data points in Figure S8 and Figure 5 in CAMS and SUST stations, which further trigger different R values.

16. Line 230. Please explain and justify this methodological choice in more detail.

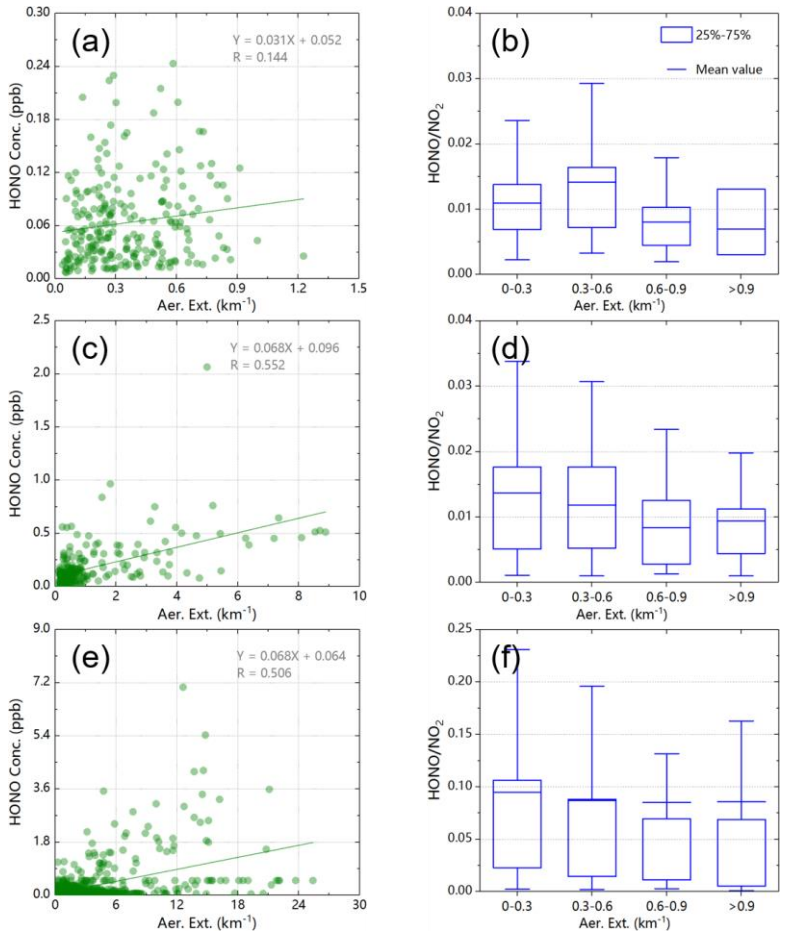
Re: Thanks for your great comments. The reason why we selected the highest values instead of mean or median values is that the variation of highest values can display an overall varying range of spots. Nearly all the data spots are below the average highest values. And the area confined by highest values and x-axis can reflect concentration levels of data spots to some extent. For example, we found that the area determined by average highest value and x-axis was much larger in the ship-based campaign than in CAMS and SUST (Fig. 6), which indicated that the varying range of HONO/NO2 ratio was much larger in sea cases than in inland cases. To eliminate the influence of other factors, we took the average of six highest $HONO/NO_2$ instead of the highest ones. We revised the method descriptions as follows.
"The highest values can represent varying range of data in each interval and reveal concentration levels of data distribution. To eliminate the influence of other factors, the average of the six highest $HONO/NO_2$ in each 10% RH interval is calculated to reflect the distribution range of data in each interval (Liu et al., 2019). The dependence of the averaged top-6 $HONO/NO_2$ on RH reveal an overall variation tendency of $HONO/NO_2$ against RH."

17. Line 231. Define "turning points" for the reader.
Re: Thanks for your great comments.
"We found the RH turning points in inland (CAMS) and coastal (SUST) cases are all ~65% (60-70%)." -> "In the inland (CAMS) and coastal (SUST) cases, the RH turning points are both ~65% (60–70%), where increasing trend switches to decreasing tendency."

18. Lines 243 to 244. Please explain and justify the methodological choice of using the average of the six highest values.
Re: Thanks for your great comments. Given that the method we used is the same with Section 3.2.1, we simplified our descriptions here.
"Similar to the scatter plots of $HONO/NO_2$ against RH, we also adopted the averaged top-6 $HONO/NO_2$ values in each 5℃ interval to represent a general variation tendency of $HONO/NO_2$ against temperature."

19. Line 252. Does "landing winds" mean a land breeze?
Re: Yes, we have changed our descriptions here.
"Moreover, we found that the appearance of $HONO/NO_2$ peak values under lower temperature (14.0-17.0℃) usually accompanied by landing wind." -> "Furthermore, we found that the appearance of $HONO/NO_2$ high values under lower temperature (14.0–17.0℃) was usually accompanied by land breeze."

20. Lines 257-260. Given that the R squared of the inland correlation is so small (<0.1), the fitted slope cannot be reliably interpreted (has no statistically significant meeting).
Re: Thanks for your great comments. We have deleted the descriptions based on the correlation results of R values < 0.1, and replaced the original figure with box plots of $HONO/NO_2$ ratio under different aerosol extinction coefficient conditions as follows.



21. Lines 263 to 266. Showing the averages and standard deviations of the ratios and aerosol extinctions in a bar chart, perhaps in the supplementary section, would help the reader. This comparison will also help to determine whether the average values are statistically significantly different based on the standard deviations, which is important to justify your conclusions.

Re: Thanks for your great comments. We have supplemented standard deviations behind the average values and added a bar chart in Figure 8 as follows.

Main text: "Additionally, we found the averaged values of $HONO/NO_2$ were 0.011 ±0.004, 0.014 ±0.006, 0.008 ±0.003, and 0.007 ±0.003 when aerosol extinctions are 0–0.3, 0.3–0.6, 0.6–0.9 and > 0.9 km^{-1} in the inland case, respectively (Figure 8(b)). As shown in Figure 8, the high values of $HONO/NO_2$ were mainly under aerosol extinction being less than 1.0 km^{-1} with averaged values of 0.012 ±0.006 and 0.090 ±0.004 in the coastal and sea cases, respectively."

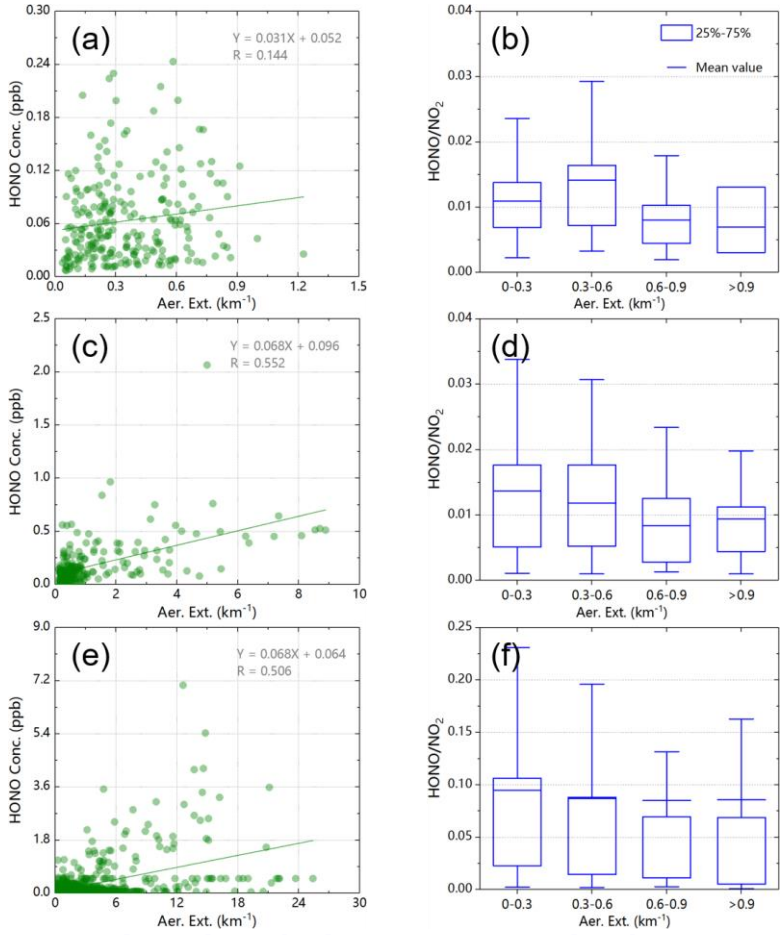


Figure 8. (a), (c), and (e) show the linear regression plots between surface aerosol extinction and HONO concentrations in CAMS, SUST and the ship-based campaign, respectively. Plots (b), (d), and (f) depicts the HONO/NO_2 ratio distribution under different aerosol extinction coefficient conditions in CAMS, SUST and the ship-based campaign.

22. Line 373. The sentence is vague. Consider revising to something like, "when peak AOD and NO2 conditions were observed, enhanced HONO were observed, but the reverse was not always the case."

Re: Thanks for your great comments.

"HONO always appeared under high AOD and NO_2 conditions." -> "When peak AOD and NO_2 conditions were observed, enhanced HONO were observed, although the reverse was not always the case."

23. Lines 374 to 376. Consider changing "to remove the primary HONO source" to "to quantify the contribution of the primary HONO source to the total production of HONO."

Re: Thanks for your great comments.

"the emission rates of $\Delta HONO / \Delta NO_x$ in sea, inland and coastal areas were calculated with values of 0.46±0.31%, 0.82±0.34%, and 0.79±0.31% to remove the primary HONO source" -> "To understand the impacts of RH, temperature, and aerosol on the heterogeneous reaction of NO_2 to produce HONO, the emission ratios of ΔHONO/ΔNOx were calculated to quantify the contribution of the primary HONO source to the total production of HONO."

24. Line 552. Figure 12 c). Suggest reducing the maximum value on the colour bar for the HONO concentrations to make the enhanced periods easier to see.

Re: Thanks for your great comments. Given that the demonstration based on Figure 12c is about HONO variation trend, we have added a line plot (Figure S12) to display the diurnal variation of HONO. We kept the same maximum value on the color bar among Figure 12, 13 and 14. In this way, it is more convenient to do a comparison of trace gas concentration levels on different days.

Main text: "The HONO was mainly distributed near the surface with a mean concentration of 0.07 ppb, and the two peaks were found in the early morning (averaged 0.15 ppb) and at 12:15 (averaged 0.11 ppb), respectively (Figure S12)."

Supplementary materials:

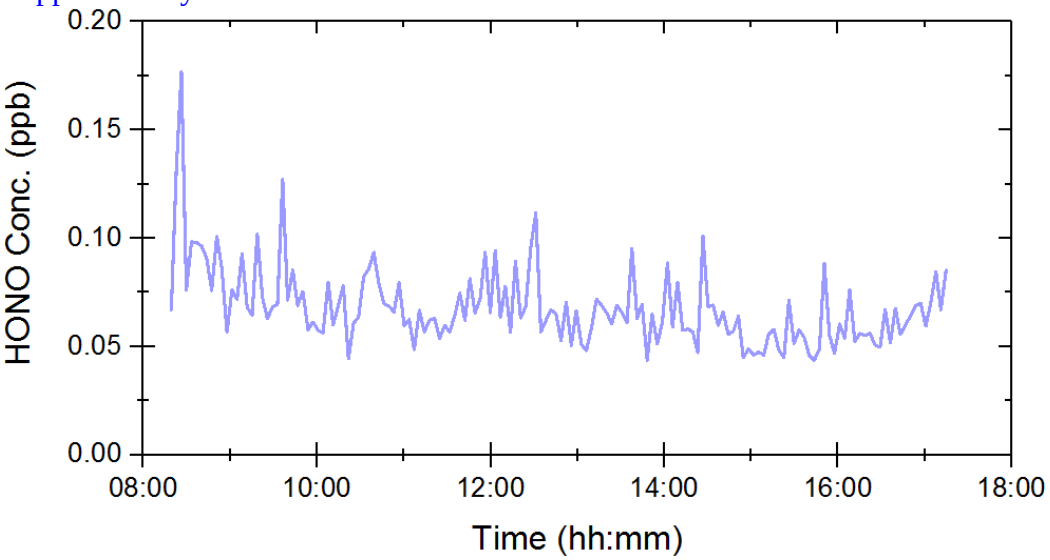


Figure S12. Time series of HONO at bottom layer on 20 April 2018.

References

Hendrick, F., Müller, J. F., Clémer, K., Wang, P., De Mazière, M., Fayt, C., Gielen, C., Hermans, C., Ma, J. Z., Pinardi, G., Stavrakou, T., Vlemmix, T., and Van Roozendael, M.: Four years of ground-based MAX-DOAS observations of HONO and NO2 in the Beijing area, Atmospheric Chemistry and Physics, 14, 765-781, 10.5194/acp-14-765-2014, 2014.

Xing, C., Liu, C., Wang, S., Chan, K. L., Gao, Y., Huang, X., Su, W., Zhang, C., Dong, Y., Fan, G., Zhang, T., Chen, Z., Hu, Q., Su, H., Xie, Z., and Liu, J.: Observations of the vertical distributions of summertime atmospheric pollutants and the corresponding ozone production in Shanghai, China, Atmospheric Chemistry and Physics, 17, 14275-14289, 10.5194/acp-17-14275-2017, 2017.