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 Prof. Kleffmann's comments. We have addressed his comments on a point-to-point basis as below for consideration, where the comments are cited in ***black****, and the responses are in* ***blue****.*

In the manuscript by Xing et al. MAX-DOAS measurements during ship cruises and on two land stations (inland and coast) were used to measure vertical gradients of HONO and NO_2 to identify potential source mechanisms. Gradient measurements are of significant importance to distinguish between near ground (e.g. direct emissions, heterogeneous NO_2 conversion, etc.) and volume sources (e.g. on particles) of HONO. Only when the vertical HONO structure is known, the impact of HONO on the oxidation capacity of the whole boundary layer can be described, in contrast to typical near surface measurements by in-situ instruments, which overweight the contribution of HONO. Also, when using a path averaging spectroscopic method the risk of overestimation of HONO levels by interferences and sampling artefacts in the instrument's inlets are minimized. Thus, such measurements are of general high importance.
However, I could not follow all the evaluations and arguments in the manuscript caused by missing information. The following comments could be considered to improve the manuscript.

Major comments:
1) Section 2.1: Missing information to CAMS and SUST sites:
Besides the ship measurements, MAX-DOAS measurements were also performed in parallel in two stations, which were defined as "inland" (CAMS) and "coastal" (SUST). Here I am missing more information to both sites. Especially, where are they? E.g. for the Chinese Academy of Meteorological Science (CAMS) I found Beijing (?), which would be far away from the ship measurements and would make any comparison highly uncertain…
Re: Thanks for your great comments.
As shown in Figure R1, Chinese Academy of Meteorological Sciences (CAMS) was located in the urban of Beijing (116.32^{o}E, 39.94^{o}N), and South University of Science and Technology (SUST) was located in Shenzhen (114.00^{o}E, 22.60^{o}N). These two MAX-DOAS stations were selected as inland and coastal cases to further understand the impacts of relative humidity (RH), temperature, and solar radiation intensity (SRI) on the heterogeneous reaction of NO_2 to form HONO in different scenes.
In order to illustrate the representativeness of CAMS as an inland scene, we selected another MAX-DOAS station (HNU: Huaibei Normal University) shown in Figure R1. Considering that there was no observation data at this station in 2018, we selected HONO and NO_2 data at the same time as ship based MAX-DOAS measurements in 2019. Moreover, we only analyzed the vertical distribution of HONO/NO_2 in HNU, due to its lack of meteorological data. As shown in Figure R2, we could find that

HONO/NO_2 decreased with the increase of height, which was the same as its performance in CAMS. Wang et al. (2020) and Meng et al. (2022) also reported the conclusion that HONO/NO_2 decreased with the increase of height. Therefore, CAMS station can represent inland scene to some extent, although it was far from the cruise route.

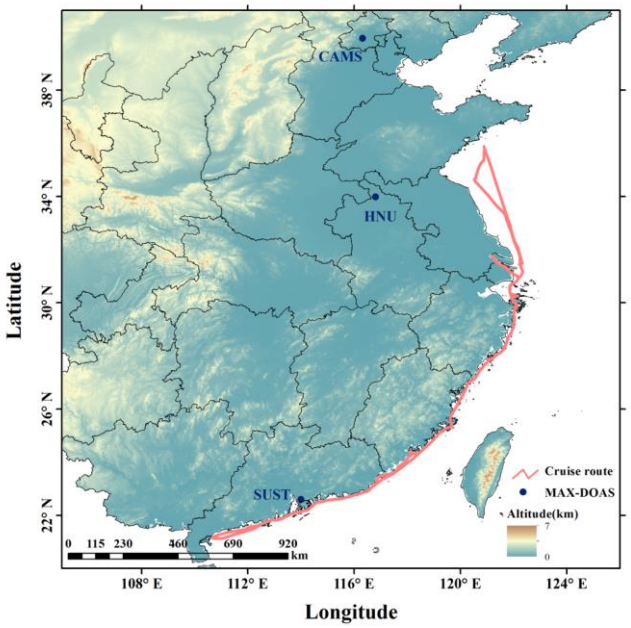

Figure R1. Cruise route and MAX-DOAS stations (CAMS, HNU and SUST).

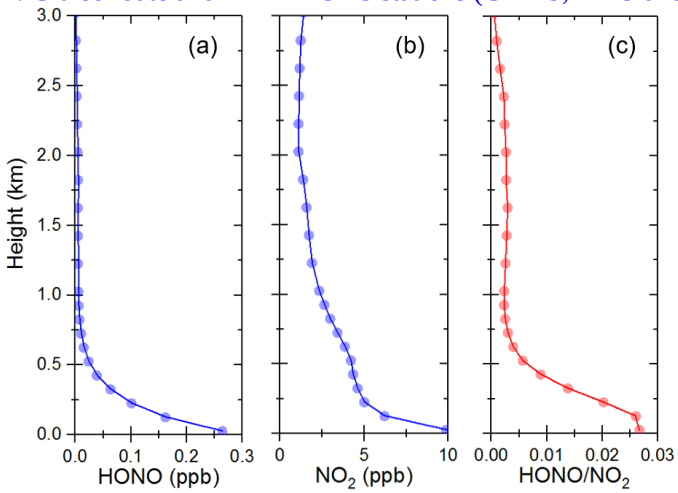

Figure R2. Vertical profiles of (a) HONO, (b) NO_2, and (c) HONO/NO_2 at HNU station from 19 April to 16 May 2019.

2) Sea- vs. land-oriented measurements:

The ship data was divided in sea- and land-oriented measurements. But isn't that both sea data? To answer this question, two important information are missing: a) How far away were the ship tracks on average from the coastline? b) what is the typical distance for the light-path of the MAX-DOAS (only the horizontal vector is of importance)? I expect that the distance of the ship from the coast (some km?) was larger than the "horizontal view" of the instrument (horizontal distance between the average scattering point and the instrument). From my experience for Chinese conditions the visibility if often significantly smaller than 1 km… In this case the instrument is only evaluating sea influenced air masses and the observed differences

reflect only some undefined horizontal gradient between sea and land, but not any "sea" of "land" data.

Re: Thanks for your great comments.

The average distance between ship and coastline was 2-20 km during the observation. The effective optical path *L* was calculated using following equation:

$$L = SCD_{O_4} / C_{O_4}$$

Where, SCD_{O_4} was the slant column density of O_4, C_{O_4} was the concentration of O_4.

The average *L* of this observation was 2-5 km. Indeed, *L* was less than the distance between ship and coastline. Moreover, we only selected data observed during clear days with visibility > 10.0 km.

Sea-oriented and land-oriented measurements can reflect the air masses affected by sea and land to some extent, respectively. Figure 3 also reported that the concentrations of aerosol, NO_2 and HONO in land-oriented measurements were all larger than that in sea-oriented measurements, considering their more obvious land sources.

In the manuscript, we modified the expression using "land-oriented measurements" and "sea-oriented measurements".

3) Direct HONO/NO_x emission ratio

In section 3.2. it seems that HONO/NO_x ratios from direct emission were determined by the measurement data for CAMS and SUST. However, it is unclear how this has been done? In the present study, only daytime data could be used (light source of the MAX-DOAS = sun…). But one filter to determine the HONO/NO_x ratio of direct emissions from field data - besides others - is to use only night-time data, caused by the fast photolysis of HONO!? In addition, because of strong vertical gradients and the vertical resolution of the MAX-DOAS the combined use of path averaged HONO and NO_2 data in comparison to in-situ NO ground data cannot be recommended (apples and oranges…). The method used is completely unclear and should be further explained. E.g. how was the direct emission ratio of 0.46% (line 216) of Sun et al. considered ("used to understand…")?

Re: Thanks for your great comments.

The MAX-DOAS measurements could be influenced by the exhaust from the measurement ship. Therefore, the data contaminated by the exhaust were filtered out. As shown in Figure R3, the direction and speed of the plume exhausted from the ship depends on the ship direction/speed and the true wind speed/direction. Individual measurements taken under unfavorable plume directions (plume directions between 45 and 135◦ with respect to the heading of the ship) were discarded. *HONO/NO_x* ratios from direct emission were determined by the measurement data for CAMS and SUST. We derived the emitted *HONO/NO_x* ratio referring to the reports in Xu et al. (2015), Liu et al. (2018) and Xing et al. (2021). 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°<SZA<75°. We put above criteria in the revised supplyment.

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°. We usually selected data with 70°<SZA<75° 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}.

Surface NO_2 was extracted from the retrieved NO_2 vertical profiles. As shown in Figure R4, the correlation coefficient (R) between surface NO_2 retrieved from MAX-DOAS and in situ NO_2 in five stations was large than 0.7 (Song et al., 2022). Therefore, we think that 0-100 m NO_2 retrieved from MAX-DOAS measurements can characterize ground surface NO_2. Moreover, Ryan et al. (2018) also used data retrieved from MAX-DOAS successfully revealed the $HONO/NO_x$ ratios from direct emission. The key problem here was how to improve the data accuracy of MAX-DOAS in the future.

For Sun et al. (2020), the detailed selection criteria of ship plumes include (a) only the data when the vessel stopped and the plume moved through the optical path were considered; and (b) concentration spikes of $HONO$ and NO_x as well as reduction in O_3 concentrations were observed.

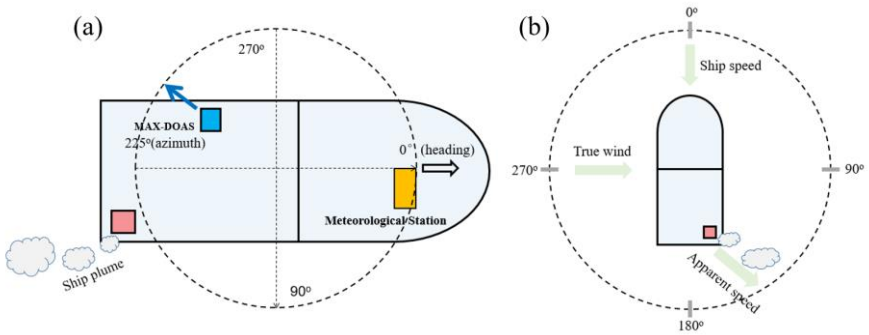

Figure R3. (a) Illustration of the MAX-DOAS setup location on the measurement ship. The red rectangle indicates the ship's exhaust. The blue rectangle represents the MAX-DOAS instrument. The blue rectangle represents the meteorological station. (b) The apparent speed and direction of plume.

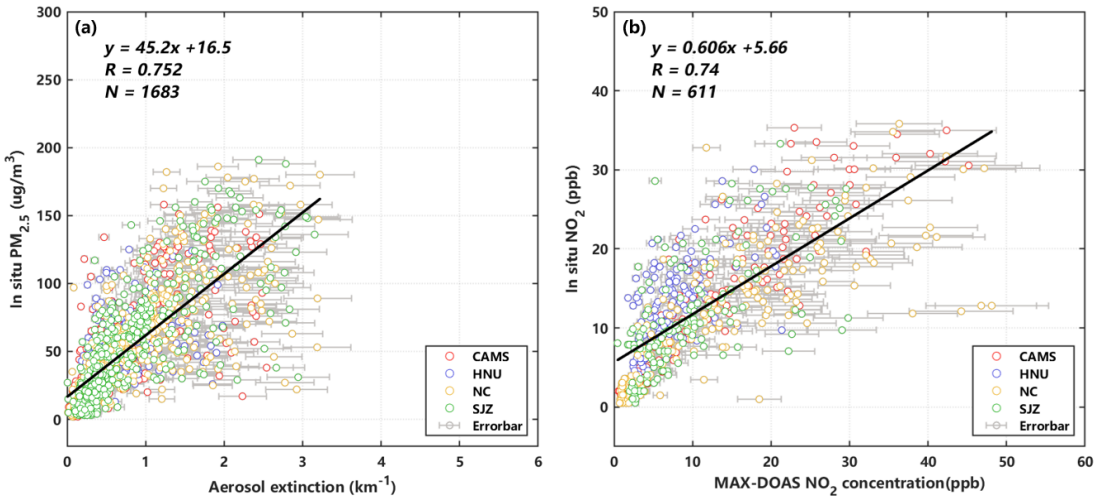

Figure R4. (a) Correlation analysis of in situ measured $PM_{2.5}$ and surface AECs (0–100 m) retrieved from CAMS, HNU, NC, and SJZ MAX-DOAS stations from January to March, 2021 and (b) their corresponding NO_2 comparative results. The black line denotes the linear least-squares fit to the data; *R* denotes Pearson correlation coefficient; *N* denotes the number of valid data. (Song et al., 2022)

4) Unrealistic $HONO/NO_x$ data:

If the HONO/NOx ratio for direct emissions of 0.82 % (CAMS) and 0.79 % (SUST) are true, then the slopes of all HONO against NO_2 data shown in Fig. 5 (a) 0.8 % for CAMS and b) 0.5 % for SUST) are not possible. Even if one assumes the absence of any NO in the atmosphere (very unreasonable) the slopes when using all data should

be by definition larger than only the direct emission ratio!? Typically, that should be a few % for field data (cf. ratio of the average ship data of ca. 2.5 %, which I get from the data in lines 191-192) for which 0.8 % (lower limit during daytime, see below) may be direct emissions. But here for SUST all data show a lower $HONO/NO_2$ ratio (and the $HONO/NO_x$ ratio would be even much lower…) than the direct emission ratio. Please check the data.

In addition, during daytime a measured $HONO/NO_x$ ratio (e.g. from sharp plumes) will be lower than what is directly emitted. This can be explained by the different lifetimes of HONO (10-20 min during daytime) and NO_2 (typically some hours). Thus, depending on the time between emission and measurements the contribution of direct emitted HONO will decrease (this is the reason why the "night-time filter" is used to measure direct emission from field data…). For details I recommend the paper by Xue et al. (https://doi.org/10.5194/acp-22-3149-2022).

Re: Thanks for your great comments.

As shown in Figure R5, we calculated the *HONO/NO₂* ratios in CAMS, SUST and the cruise during the observation. The average *HONO/NO₂* ratios in CAMS and SUST were 0.012 and 0.014, respectively, which were significantly higher than corresponding fitting slopes and the *HONO/NO₂* emission ratios. The average *HONO/NO₂* ratios during the cruise were 0.20-0.25. We put this figure in the revised supplyment.

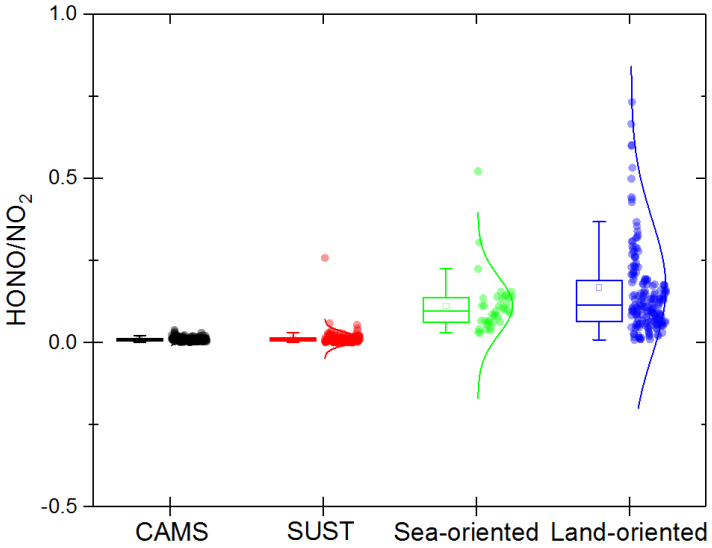

Figure R5. $HONO/NO_2$ ratios in CAMS, SUST and the cruise.

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°<SZA<75°. As we all know, 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°. In order to reduce the influence of fast photolysis of *HONO* and NO_2, we usually selected data with 70°<SZA<75° 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}. We also have learned the paper of Xue et al. (2022).

5) Unrealistic $HONO/NO_2$ gradient data:

In figures 9-11 vertical gradient data of the $HONO/NO_2$ ratio are shown. Here increasing ratios are observed with altitude, which is in contrast to most gradient data,

which I know (cf. e.g. our gradient data on a 190 m tall tower, Kleffmann et al., 2003 doi: 10.1016/S1352-2310(03)00242-5). While this may be explained by any unusual chemistry over sea surfaces, the absolute numbers of the HONO/NO_2 at higher altitude of up to 45 % (see Fig. 10) are impossible, independent of how strong any HONO source – e.g. particle nitrate photolysis – may be. The photolysis of HONO is a source of NO. In a typical atmosphere for which [O_3]>[NO_x] this is quickly converted to NO_2. Since in higher layers in a well-mixed atmosphere a PSS can be assumed (far away from any direct sources) the maximum HONO/NO_2 ratio is given by the ratio of the lifetimes of both molecules. For HONO this is around 10 min at noon (check for J(HONO)), while for NO_2 this is mainly limited by its reaction with the OH radical during daytime (the Leighton chemistry will not play a role here). Assuming a high OH concentration of 10^7 cm^{-3} at 1 km altitude a lifetime of ca. 3 h can be calculated. Thus, a maximum HONO/NO_2 ratio of ca. 6 % should result under steady state conditions. If HONO is measured close to a source, e.g. in near ground measurements in a step vertical gradient, higher HONO/NO_2 ratios are possible (= no PSS…). But in a homogeneous mixed atmosphere at 1 km altitude (see figures 9-11) such high HONO/NO2 data is impossible. Please check.

Re: Thanks for your great comments.

As shown in Figure 10, the *HONO/NO₂* ratio in CAMS was decreasing with the increase of height under 200 m with aerosol extinction coefficient less than 0.2 km^{-1}. The average *HONO/NO₂* in CAMS under 200 m was 0.015 during the campaign, which was within the range of *HONO/NO₂* (0.0-0.07) in previous studies (Kleffmann et al., 2003; Meng et al., 2020). Moreover, Zhang et al. (2020) also reported that the *HONO/NO₂* ratio in Beijing increased with the increase of height under 200 m in haze days. Figure 11 also told us that the *HONO/NO₂* ratio in CAMS also increased with the increase of height under the condition of extinction coefficient larger than 0.7 km^{-1}.

In order to understand the accuracy of MAX-DOAS data, we analyzed the retrieval quality of MAX-DOAS data described in Figure 10-11 as following.

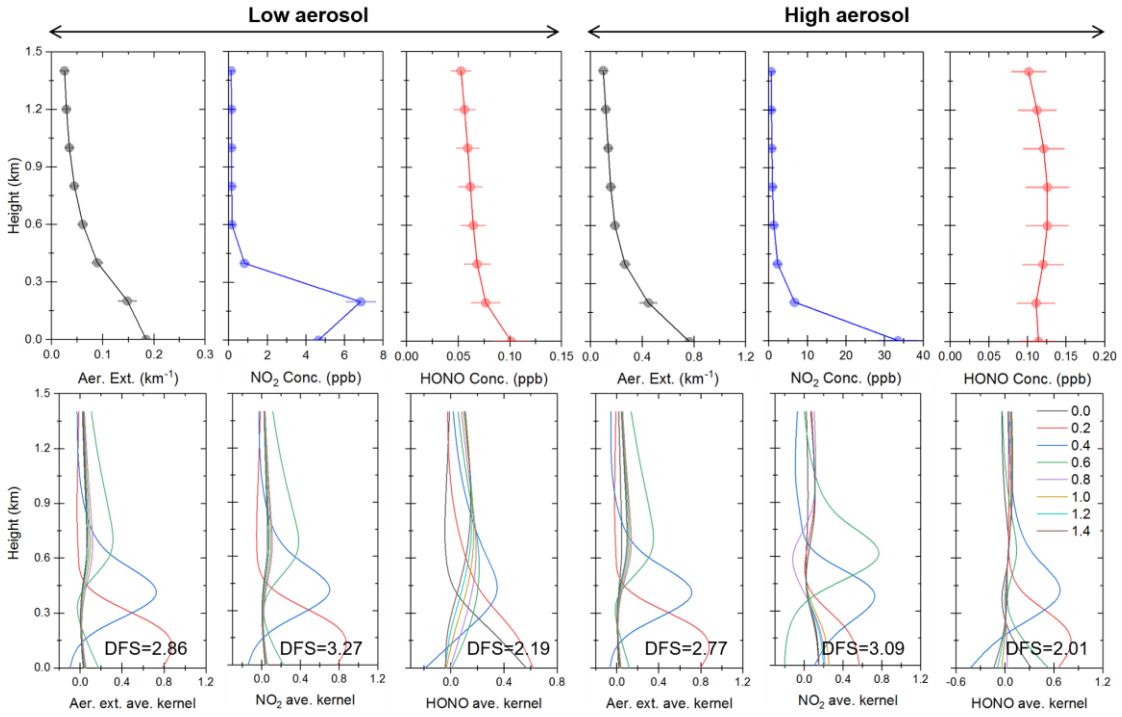

Figure R6. The top row presented the vertical profiles and errors of aerosol, NO_2 and *HONO* under low aerosol and high aerosol conditions. The bottom row showed the corresponding retrieved averaging kernels.

Figure R6 told us that the data quality was reliable. This section was put into the revised supplyment.

About the high HONO/NO_2 ratio (~0.45) during the cruise observation (Figure 10):

We could find that there was an obvious mutation in *HONO/NO_2* ratio at about 0.5 km. The HONO air mass above 0.5 km maybe detected during this process. As shown in Figure R7, we plotted all the *HONO/NO_2* ratios during the cruise observation. We also could find the increase of *HONO/NO_2* with the increase of height. This figure was put into the revised supplyment.

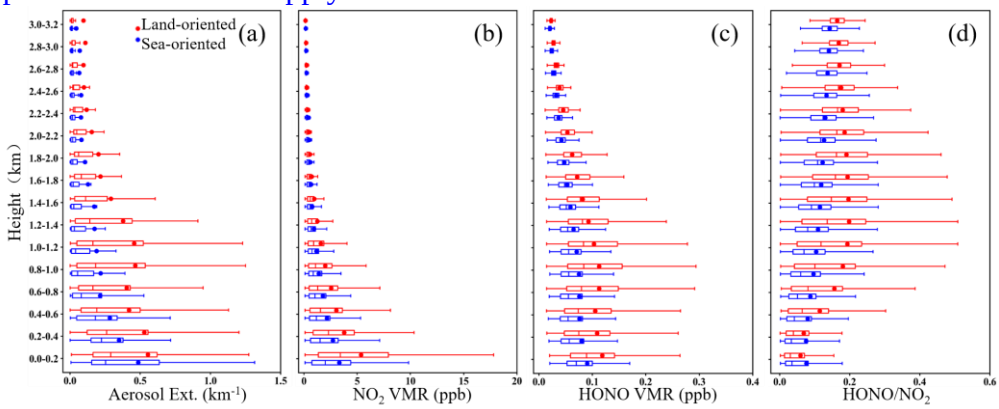

Figure R7. Vertical profiles of (a) aerosol extinction, (b) NO_2, (c) HONO, and HONO/NO_2 ratios during the cruise observation.

Minor comments in the order of the manuscript:

Line 37-38: There are several "heterogeneous reactions of NO_2". Here the authors should distinguish between slower nighttime conversion (NO_2+H_2O and NO_2+organic) and daytime sources (NO_2+organic + light, see Stemmler et al., 2006; or NO_2 +TiO_2+light = photocatalysis). Otherwise some arguments of the authors (with solar radiation, see below) are unclear.

Re: Thanks for your great comments.

We have rewritten this sentence as following:

"the known sources of HONO mainly include direct emissions from vehicles, ships, biomass burning and soil, the homogeneous reaction of NO and OH radicals, the nighttime and daytime heterogeneous reaction of NO_2 on aerosols, vegetation, ground and other types of surfaces, and the photolysis of nitrate particles (NO_3^-) (Stemmer et al., 2006; Indarto et al., 2012; Wang et al., 2015)."

Line 51-53, general comment to this section, but also to the author's own evaluations: These simple correlation studies always bear the risk of a misinterpretation of the results. Typically, trace gases which are emitted or formed near to the ground will anyhow correlate caused by the variable mixing layer height. The is mainly modulated by diurnal surface temperature variation which has also an effect on the relative humidity. Thus, e.g. at the end of the night the temperature and mixing height are low, while the relative humidity is high. Caused by the resulting high S/V ratio under these conditions, heterogeneous HONO formation is faster and the HONO/NO_x ratio will correlate with the humidity, without any necessary mechanistic link (see also correlation of Radon with HONO…). Also, often at very high humidity the HONO/NO_x ratio is again decreasing with humidity. This is typically explained by

uptake on very humid surfaces. However, the highest relative humidity is often observed close before sunrise, when direct emissions start to increase. Thus, the high HONO/NO_x air masses from slow nighttime sources (typically 5 %) are "diluted" by fresh low HONO/NO_x emissions (around 1%), leading to the decreasing HONO/NO_x ratios at high humidity. Thus, the authors should highlight (and later consider for their own evaluation…) that simple correlation analysis may lead to artificial correlations and misleading conclusions.

Re: Thanks for your great comments. This suggested that more detailed process analysis and quantitative analysis in addition to linear regression analysis should be valued in the future. In this process, with the help of multiple models and cooperation with superior teams, data advantages can be better played.

We have rewritten these sentences as following:

Previous works always used the linear regression relationship between HONO/NO_2 and above parameters to characterize the influence of these parameters on the formation of HONO through the heterogeneous reaction of NO_2. Although this kind of simple linear regression method may lead to artificial correlations and misleading conclusions, considering the vertical evolution of atmospheric parameters. Wen et al. (2019) found that the increased temperature could promote the heterogeneous reaction of NO_2 to form HONO in sea conditions. The generation rate of HONO could increase rapidly, when the temperature is greater than 20 ℃. Gil et al. (2019) found that the HONO formed from the heterogeneous reaction of NO_2 will increase along with the increase of RH when RH is less than 80% in a case of land park using deep learning forced by measurement results. Fu et al. (2019) reported that RH and SRI are the main parameters driving the heterogeneous reaction of NO_2 to form HONO in Pearl River Delta, and it contributes 72% of the total source of HONO. Cui et al. (2019) found that the potential of heterogeneous reaction of NO_2 to form HONO will increase with the increase of particle concentration and the specific surface area of single particle in coastal cities.

Line 77-85: With respect to the main topic of the manuscript, I would expect a more extended summary of the existing gradient data (from towers, and MAX-DOAS), which is normally very different to the present results (see major comment 6).

Re: Thanks for your great comments.

Taking tower and aircraft as platforms, these techniques performed to measure HONO vertical profiles, and 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. Moreover, they also reported that the HONO/NO_2 ratios usually decreased with the increase of height under 200m at inland and coastal areas. However, the cost of above techniques used to measure HONO vertical profiles was too high, and the real-time and continuous measurement cannot be realized. Multi-axis differential optical absorption spectroscopy (MAX-DOAS), as a ground-based ultra-hyperspectral remote sensing technology, has been widely used for vertical observation of atmospheric pollutants in the past two decades. In the past five years, several researchers have carried out campaigns based on MAX-DOAS to measure the vertical profile of HONO in inland and coastal areas, and revealed their vertical characteristics, sources and the contribution to atmospheric oxidation at different height layers (Garcia-Nieto et al., 2018; Ryan et al., 2018; Wang et al., 2020;

Xing et al., 2021; Xu et al., 2021; He et al., 2023). There were few studies on the sources of HONO at different height layers in sea conditions. In this study, it will be the first time to use MAX-DOAS to study the spatiotemporal distribution and the sources of HONO along the Chinese coastline, and to learn the differences of the HONO formed from the heterogeneous reaction of NO_2 in different height layers and land-sea scenes.

Line 187-189: This sentence could make sense only if a photolytic NO_2 conversion process is considered (see above). However, even for a photolytic NO_2 conversion process which was found to correlate with $J(NO_2)$ in lab studies (see Stemmler et al., 2006), the steady state $HONO/NO_2$ ratio would not change with variable solar radiation, since both, J(HONO) (sink) and $J(NO_2)$ (source) show a linear correlation. Thus, the argument is not valid.

Re: Thanks for your great comments. The following sentence and Figure S2 were removed in the revised manuscript and supplyment, respectively.

"On the other hand, the solar radiation intensity in this day (12 May, 2018) was significantly lower than other days (Fig. S2), and this weather condition was not conductive to the HONO formation through the heterogeneous reaction of NO_2."

Lines 191-192 and 205: Here very different $HONO/NO_2$ ratios are specified for the same (?) ship data? From the data in lines 191-192 I get values of 2.7 % and 2.4 % ("total averaged"), while in line 205 45 % are mentioned for the "average value"? Check data and/or explain differences.

Re: Thanks for your great comments. 0.027 and 0.024 were the average values of $HONO/NO_2$ at sea-oriented and land-oriented measurements during the whole campaign. 0.45 was the average value of $HONO/NO_2$ on 02, 12 and 14 May. The sentences have been rewritten as following:

"The surface concentration of NO_2 and HONO were extracted from their corresponding vertical profiles. As shown in Figure 3, the total averaged near-surface NO_2 concentrations under sea-oriented and land-oriented measurements were 8.46 and 11.31 ppb, respectively. The total averaged near-surface HONO concentrations were 0.23 and 0.27 ppb under sea-oriented and land-oriented measurements. Previous studies reported that vehicle and ship emissions were the main primary HONO sources on land and sea, respectively, and NO_2 heterogeneous reaction on the surfaces of ground, sea, vegetation and aerosol were the HONO important secondary sources (Liu et al., 2021). They also found that the surface HONO concentration under sea case was lower than that under land case, especially in the morning and evening (Yang et al., 2021). Figure 4 showed the time series of AOD, the surface concentrations of NO_2 and HONO, and the surface $HONO/NO_2$ during the whole campaign. We could find the time series of AOD and NO_2 were similar. The high AOD and NO_2 usually appeared in busy shipping channels and ports, and the obvious high-value areas were the coast of the Yangtze River Delta, the Taiwan Strait, Xiamen port, Zhanjiang port and Qingdao port (with mean AOD of 1.28 and mean NO_2 of 18.90 ppb). HONO always appeared under high AOD and NO_2 conditions, however, high AOD and NO_2 were not necessarily accompanied with high HONO concentration. This was because the heterogeneous formation of HONO requiring suitable meteorological conditions (i.e., RH and temperature) in addition to its

precursor (NO_2) and the reaction surface (aerosol) (Liu et al., 2019). The high $HONO/NO_2$ values were found on 02, 13 and 14 May with an average value of 0.45. Moreover, we found the high values of $HONO/NO_2$ always appeared from 11:00 to 14:00 during a whole day."

Line 202: should be "high HONO concentration". A production rate (dHONO/dt) was not determined and you may have a small production rate (slope) at high HONO.
Re: Thanks for your great comments. We have rewritten this sentence as following:
"HONO always appeared under high AOD and NO_2 conditions, however, high AOD and NO_2 were not necessarily accompanied with high HONO concentration."

Line 206-207: Check again the argument (see above, sources and sink scale with radiation…).
Re: Thanks for your great comments. The following sentence was removed in the revised manuscript.
"That was due to the high production rate of HONO and the high photolysis rate of NO_2 during noontime"

Section 3.2.1: Check whether the "turning points" (especially the two in Fig. 6c) are significant or just scatter of the data? In addition, possible "artificial correlations" should be discussed, see above.
And can you explain, why only the "six highest values" are shown in Fig. 6 (red data) and not the mean/median? Is that representative or are here only outliers shown?
Re: Thanks for your great comments.
In order to eliminate the influence of other factors, the average of six highest $HONO/NO_2$ in each 10% RH interval is calculated. The bands of RH were selected to be 40-50%, 50-60%, 60-70%, 70-80%, 80-90% and 90-100% in Figure 6 (c). In order to prove whether there was possibility of artificial correlation, we selected RH intervals of 5% (40-45%, 45-50%, 50-55%, 55-60%, 60-65%, 65-70%, 70-75%, 75-80%, 80-85%, 85-90%, 90-95% and 95-100%). We used mean $HONO/NO_2$ values during this process. In Figure R8, we could also find two turning peaks appearing at ~60% and ~85% (80-90%), respectively. As reported by Cui et al. (2019), it can also be found that two similar RH turning peaks corresponding to higher $HONO/NO_2$ values from the observation data in East China Sea, although they did not clearly explain this phenomenon in their manuscript.

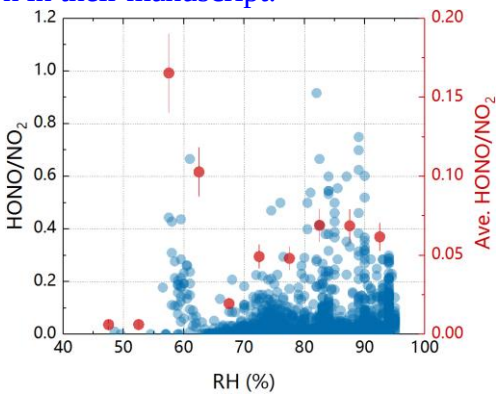

Figure R8. Scatter plots of RH and $HONO/NO_2$ ratios in the ship-based campaign.

Line 245, 246, 251: Here continuously increasing or decreasing data is shown and the highest value are specified as "peak". However, the "peak values" were not determined and could be even at lower or higher temperatures…

Re: Thanks for your great comments. We have rewritten the sentences as following:

(1) "In inland condition (CAMS), the $HONO/NO_2$ decreased along with the increase of temperature, and the highest values of $HONO/NO_2$ appeared on ~12.5℃."

(2) "However, we found that $HONO/NO_2$ increased along with the increase of temperature, and the highest values of $HONO/NO_2$ appeared with ~31.5℃ in coastal condition (SUST)."

(3) "In sea condition, the $HONO/NO_2$ increased along with the increase of temperature with a high value under ~25.0℃ when the atmospheric temperature was larger than 18.0℃, simultaneously, a ~1.9 averaged $HONO/NO_2$ high value was found under ~15.0℃ (14.0-17.0℃)."

(4) "Moreover, we found that the appearance of $HONO/NO_2$ high values under lower temperature (14.0-17.0℃) usually accompanied by landing wind."

Paragraph lines 282-295/ figures 10 and 11: What is the difference between both figures? Seems to be the same? Define two cases?

Re: Thanks for your great comments. We would like to understand the difference of the vertical evolution of $HONO/NO_2$ under inland and sea scenes under different aerosol loads. Figure 10 introduced a case with low aerosol level (<0.2 km^{-1}) but with similar vertical shape of aerosol under inland and sea scenes. Figure 11 introduced a case with relatively high aerosol level (~0.8 km^{-1}) but with similar vertical shape of aerosol under inland and sea scenes.

We have rewritten these sentences as following:

"In addition, we selected inland cases (CAMS) to learn the difference of height dependence of $HONO/NO_2$ compared with sea scenes under different aerosol loads. As shown in Figure 10, the sea and inland scenes had the similar aerosol levels (low aerosol level: < 0.2 km^{-1}) and vertical structure. Moreover, the NO_2 and HONO in sea and inland scenes had the similar vertical structure, but their concentrations in sea scene are all larger than that in inland scene. In Figure 10(d), we could find that the $HONO/NO_2$ in sea scene was obviously larger than that in inland scene above 400 m. The $HONO/NO_2$ in sea scene was about 4.5 times larger than that in inland scene especially above 600 m. As shown in Figure 11, the aerosols under sea and inland scenes were also with the similar extinction levels (relatively high level: ~0.8 km^{-1}) and vertical structure. The NO_2 concentration in sea scene was higher than that in inland scene but with a similar vertical structure. The HONO concentration in sea scene was lower than that in inland scene under 400 m, while it in sea scene was larger than that in inland scene above 400 m. In Figure 11 (d), we found the $HONO/NO_2$ in inland scene was larger than that in sea scene under 600 m, while the $HONO/NO_2$ in sea scene was about 2 times larger than that in inland scene above 600 m. Above all cases indicated that the HONO generation rate from NO_2 heterogeneous reaction in sea scene was larger than that in inland scene in higher atmospheric layers above 400-600 m. The high-altitude (> 400-600 m) atmospheric parameters in sea scene were more conductive to promote the HONO formation through the heterogeneous reaction of NO_2."

Line 315: Where is that HONO peak at 12:15 in Figure 12c? I see a stronger peak at ca. 14:15…?

Re: Thanks for your great comments. There was a HONO peak at 12:15. In order to observe the data more intuitively, we plotted the HONO concentration at bottom layer on 20 April in Figure R9. We put this figure into the revised supplyment.

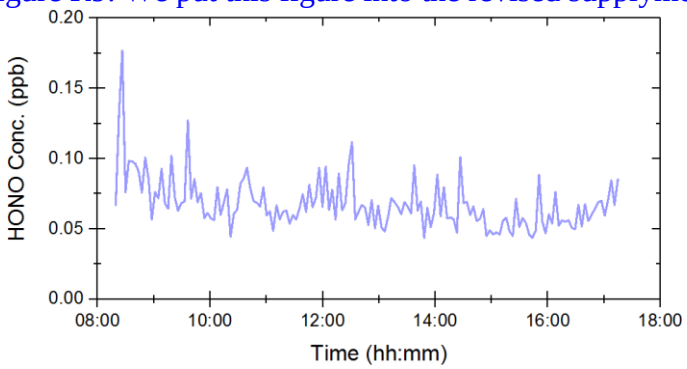

Figure R9. Time series of HONO at bottom layer on 20 April 2018.

Line 330-331: The two RH and especially the two T values are not very different to allow any conclusions to the mechanism.

Re: Thanks for your great comments. We have rewritten this sentence as following:

"The slightly increase of RH and temperature (Tem) at 14:00-16:00 (RH: ~75.0%, Tem: 23.7℃) may contribute to HONO formation through heterogeneous reaction of NO_2 on the aerosol surface than that at 09:00-11:00."

Line 343-344, Fig. 15: Not the NO_2 concentration is increasing during this period (see color code), but the layer is getting thicker.

Re: Thanks for your great comments. As shown in Figure R10, we could find that NO_2 increased under 1.0 km from 08:00 to 12:00. We put this figure into the revised supplyment.

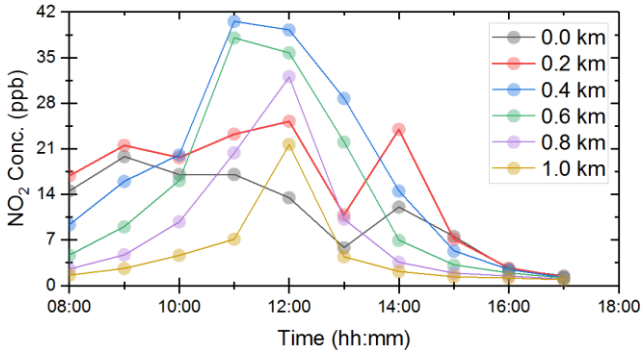

Figure R10. Time series of NO_2 at 6 layers on 03 May 2018.

Line 347-348. The peaks in HONO at ca. 9:45, 11:00, 11:45 and 12:30 in Fig. 15 are anticorrelated to NO_2 (in contrast to the statement…), which is very unusual? Check data and sentence.

Re: Thanks for your great comments. We checked the NO_2 and HONO data in this case, and the peaks of NO_2 and HONO at 0.5-1.0 km indeed appeared simultaneously from 09:45 to 13:00. We have rewritten this sentence as following:

"Several HONO peaks (> 0.2 ppb) at 0.5-1.0 km were found from 09:45 to 13:00, and the aerosol and NO_2 high values were also observed at this height layer, simultaneously."

Line 375: Should be "emission ratio".

Re: Thanks for your great comments. We have rewritten this sentence as following:

"In order to further understand the impacts of RH, temperature, and SRI on the heterogeneous reaction of NO_2 to produce HONO, the emission ratio 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."

Fig. 1: The data shown seems to be not "typical". The DSCDs in the figure are factors higher than the data described in section 3.1?

Re: Thanks for your great comments.

In section 3.1, we used VCD to depict the variation of NO_2 and HONO along the cruise route. The relationship between DSCD and VCD was $VCD = DSCD/DAMF$. In this study, a radiative transfer model SCIATRAN was used to convert SCDs of NO_2 and HONO to their tropospheric vertical column densities (VCDs). The vertical profiles of aerosol, NO_2 and HONO retrieved from MAX-DOAS, the temperature and pressure vertical profiles simulated using a dynamical-chemical model (WRF-Chem), and the geo-position data collected by GPS were introduced as inputs in SCIATRAN for the NO_2 and HONO air mass factor (AMF) calculation.

We also provided the conversion relationship between DSCD and VCD based on geometric AMF to help you to quickly quantify this relationship.

$VCD = DSCD/\left((1/\sin\alpha)-1\right)$.

α was the elevation angle. In actual observation, the real AMF (radiative transfer model based) will be larger than the geometric AMF, due to the multiple scattering effect of aerosols in the atmosphere.

Figure 3. Check the $HONO/NO_2$ data. I get 0.027 and 0.024 using the HONO (0.23 /0.27) and NO_2 (8.46/11.31) data?

Re: Thanks for your great comments. We have updated Figure 3 according to the actual observation data. The average $HONO/NO_2$ ratios in sea-oriented and land-oriented measurements should be 0.027 and 0.024, respectively.

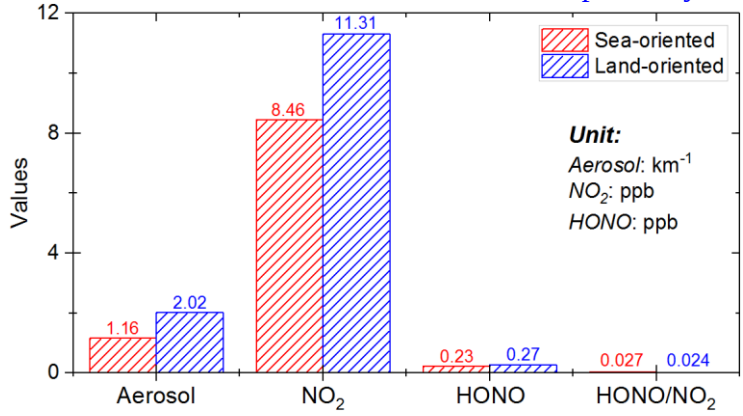

Figure 3. Averaged aerosol extinction, NO_2 concentration, HONO concentration and $HONO/NO_2$ ratio during the campaign. The red and blue boxes denoted sea-oriented and land-oriented measurements, respectively.

Figure 6: please show the red/right y-axis scaling in all figures (will be different in a) and b)).

Re: Thanks for your good suggestion. We have replotted Figure 6 as following:

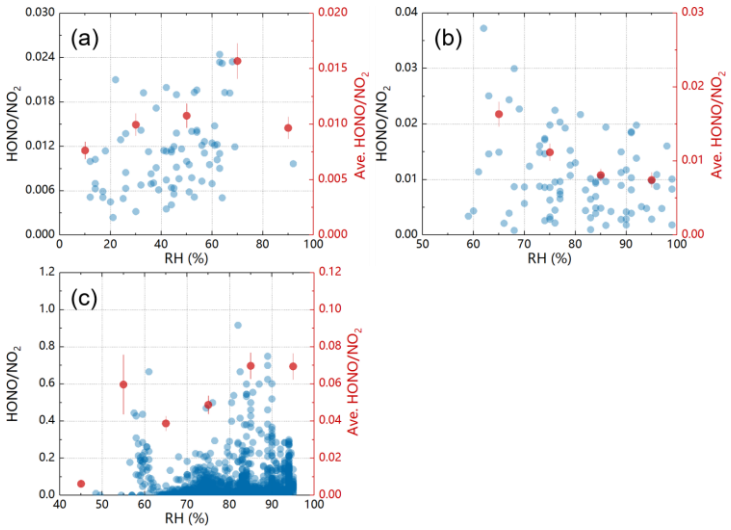

Figure 6. Scatter plots of RH and $HONO/NO_2$ ratios in (a) CAMS, (b) SUST, and (c) the ship-based campaign.

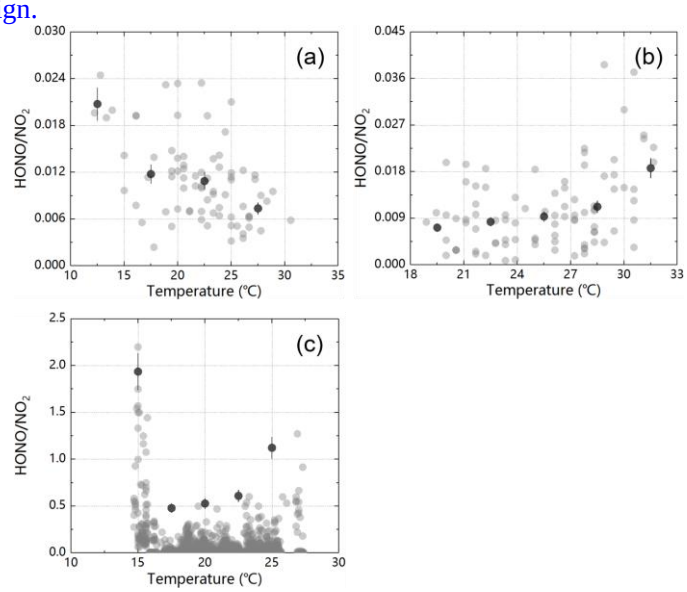

Figure 7. Scatter plots of temperature and $HONO/NO_2$ ratios in (a) CAMS, (b) SUST, and (c) this ship-based campaign.

Figure cations 10 and 11. Is “Sea” the average data or “sea/land oriented data”?

Re: Thanks for your good suggestion. The cases described in Figure 10 and Figure 11 were selected from all of the ship-based campaign, and sea-oriented and land-oriented were not distinguished. We have replotted Figure9-11 as following:

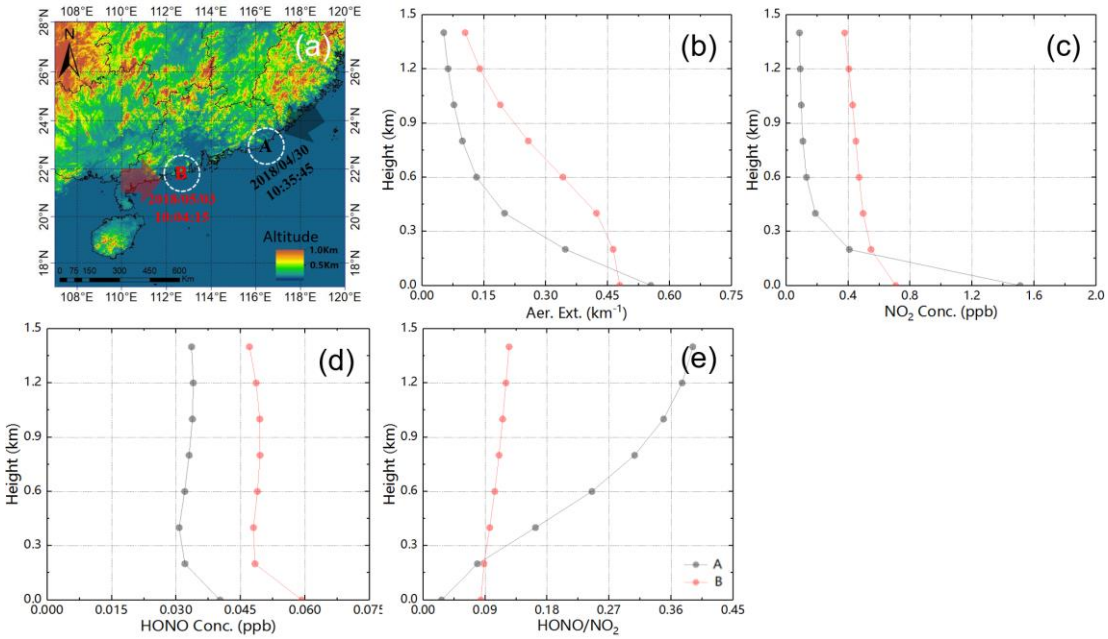

Figure 9. (a) showed two measurement points (A: black, sea-oriented with sea wind; B: red, land-oriented with land wind) during the campaign. (b)-(e) showed the vertical profiles of aerosol, NO_2, HONO, and HONO/NO_2 ratios in above two measurement points, respectively.

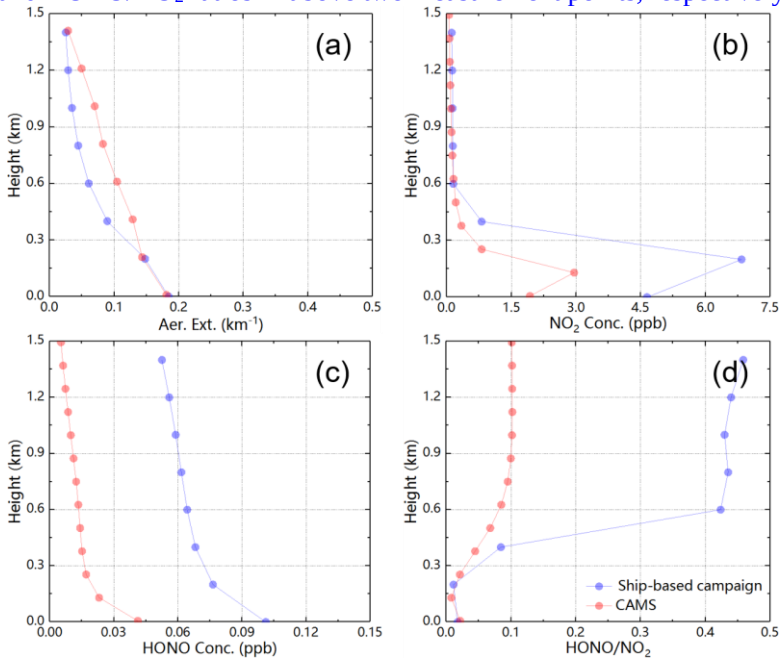

Figure 10. Vertical distributions of (a) aerosol extinction, (b) NO_2 concentration, (c) HONO concentration, and (d) HONO/NO_2 ratio. The blue and red lines represented a ship-based campaign case and a CAMS case, respectively.

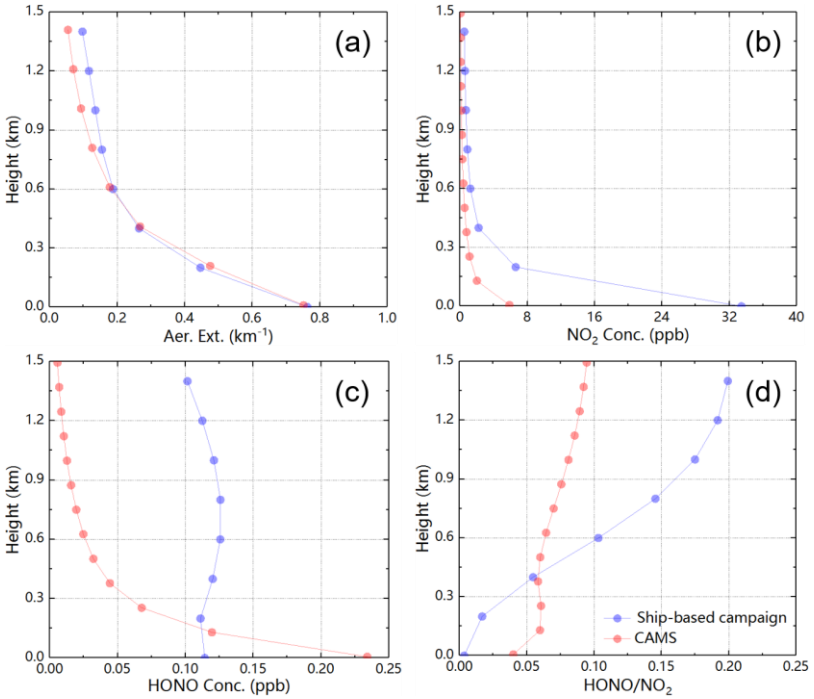

Figure 11. Vertical distributions of (a) aerosol extinction, (b) NO_2 concentration, (c) HONO concentration, and (d) HONO/NO_2 ratio. The blue and red lines represented a ship-based campaign case and a CAMS case, respectively.

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