A point-by-point response to Referee #1

We sincerely appreciate Referee #1 for the valuable comments and constructive suggestions that helped improve the quality of our manuscript. The following is a point-by-point response to address the referee's comments. The original comments are shown in *black*, and our corresponding responses are presented in *blue*. The new or modified contents in the revised manuscript are marked in *red*.

Comments from Referee #1:

This manuscript studied HONO concentrations and its sources in urban Beijing during autumn and winter of 2022. The results showed that NO_2 heterogeneous reaction on ground was the dominant HONO source. Vehicle emissions and nitrate photolysis also contributed to HONO concentrations. In general, the research is interesting, the results and discussions are sounds. Here are some technique comments need to be addressed before it can be accepted.

Response: Many thanks to Referee #1 for the valuable comments and constructive suggestions, which are significant for improving the quality of the manuscript. We carefully revised and supplemented the manuscript in response to the referee's comments on the technique comments. The following are point-by-point responses to the referee's comments.

Detailed comments:

1. L78, studied.

Response: Thanks for your valuable comments. Revision has been made as the referee suggested. Lines 80-82 in the revised manuscript:

"It provided a unique opportunity to identify HONO sources and their potential impact to secondary pollution formation in urban Beijing, which has been rarely studied in the past."

2. L114-115, the NO concentrations measured by chemiluminescence NOx analyzer is ok. But the analyzer could overestimate NO_2 concentrations due to include other oxidized nitrogen. You can calibrate the data using the method in JGR: Atmospheres, 127, e2021JD036379. https://doi.org/10.1029/2021JD036379.

Response: Thanks for your valuable comments. Revision was made as the referee suggested. We corrected CL_NO_2 (Thermo Scientific, Model 42i NOx analyzer) using interference-free CAPS_NO_2 measurements (Teledyne API-N500 NOx analyzer), and provided rigorous field comparison evidence. To improve the precision and accuracy of the NO_2 correction, we established separate daytime and nighttime linear regressions. All relevant parameters were recalculated with the corrected NO_2, and the model simulations were rerun. The correction methodology and its impacts were described in detail in the Supporting Information (see Text S1 and Figure S1).

Text S1 in the Supporting Information:

"As the most important precursor of HONO, accurate measurement of NO_2 was crucial for analyzing HONO formation. A commercial Thermo Scientific analyzer (42i) used in this study could specifically detect NO. The measurement of NO_2 was achieved by converting NO_2 to NO through a molybdenum converter. However, the chemiluminescence (CL) technique could overestimate NO_2 concentrations because of the interference of NOy. These interferences included HONO, HNO_3, HNO_4, N_2O_5, NO_3, peroxyacetyl nitrate (PANs, $RC(O)OONO_2$), organic nitrates ($RONO_2$), and peroxynitrates ($ROONO_2$) (Villena et al., 2012; Wu et al., 2022). Therefore, the NO_2 measured by the CL-NOx analyzer represented the sum of real NO_2 and these interfering species. In contrast, the commercial Teledyne API-N500 NOx analyzer was based on cavity attenuated phase shift (CAPS) technique. It could provide direct absorption measurement of NO_2 at 450 nm in the blue region of the electromagnetic spectrum, allowed fast and accurate detection of NO_2 without interference from water vapor. The only known potential interferences in the typical ambient environment were dicarbonyl compounds such as glyoxal and methylglyoxal, whose concentrations were usually much lower than NO_2 mixing ratios (Kebabian et al., 2008). Therefore, NO_2 measured by the CAPS-NOx analyzer (CAPS_NO_2) could be used to correct the NO_2 measured by the CL-NOx analyzer (CL_NO_2).

We conducted a NO_2 field campaign at the ICCAS site from September 19 to October 11, 2023, to compare the performance of the CL-NOx and CAPS-NOx analyzers. The sampling inlets of both instruments were placed at the same location, with identical sampling tube lengths, and the analyzers were housed in the same indoor environment to minimize external interference. The results showed that CAPS_NO_2 and CL_NO_2 exhibited similar temporal variations (Figure S1(a) and S1(b)). Notably, CL_NO_2 was consistently higher than CAPS_NO_2, with a more pronounced difference during the daytime. This discrepancy was mainly attributed to elevated NOy concentrations caused by enhanced photochemical reactions. Consequently, the fraction of CAPS_NO_2 in CL_NO_2 displayed a distinct diurnal pattern, being higher at night and lower during the day (Figure S1(c)), which was consistent with previous findings (Xue et al., 2022; Zhang et al., 2022c). Based on this result, we applied separate calibrations for daytime (07:00-18:00 LT) and nighttime (19:00-next 06:00 LT) data. The results indicated strong linear correlations between CAPS_NO_2 and CL_NO_2 during both periods ($R^2 = 0.96$ for daytime and $R^2 = 0.95$ for nighttime). The regression equations were "$y = 0.98x - 2.27$" for daytime and "$y = 0.99x - 2.29$" for nighttime, where y represented CAPS_NO_2 and x represented CL_NO_2 (Figure S1(d) and S1(e)). Using these relationships to correct the NO_2 data obtained in this study provided a more reasonable estimation of true NO_2 concentrations and offered a reliable basis for further analysis."

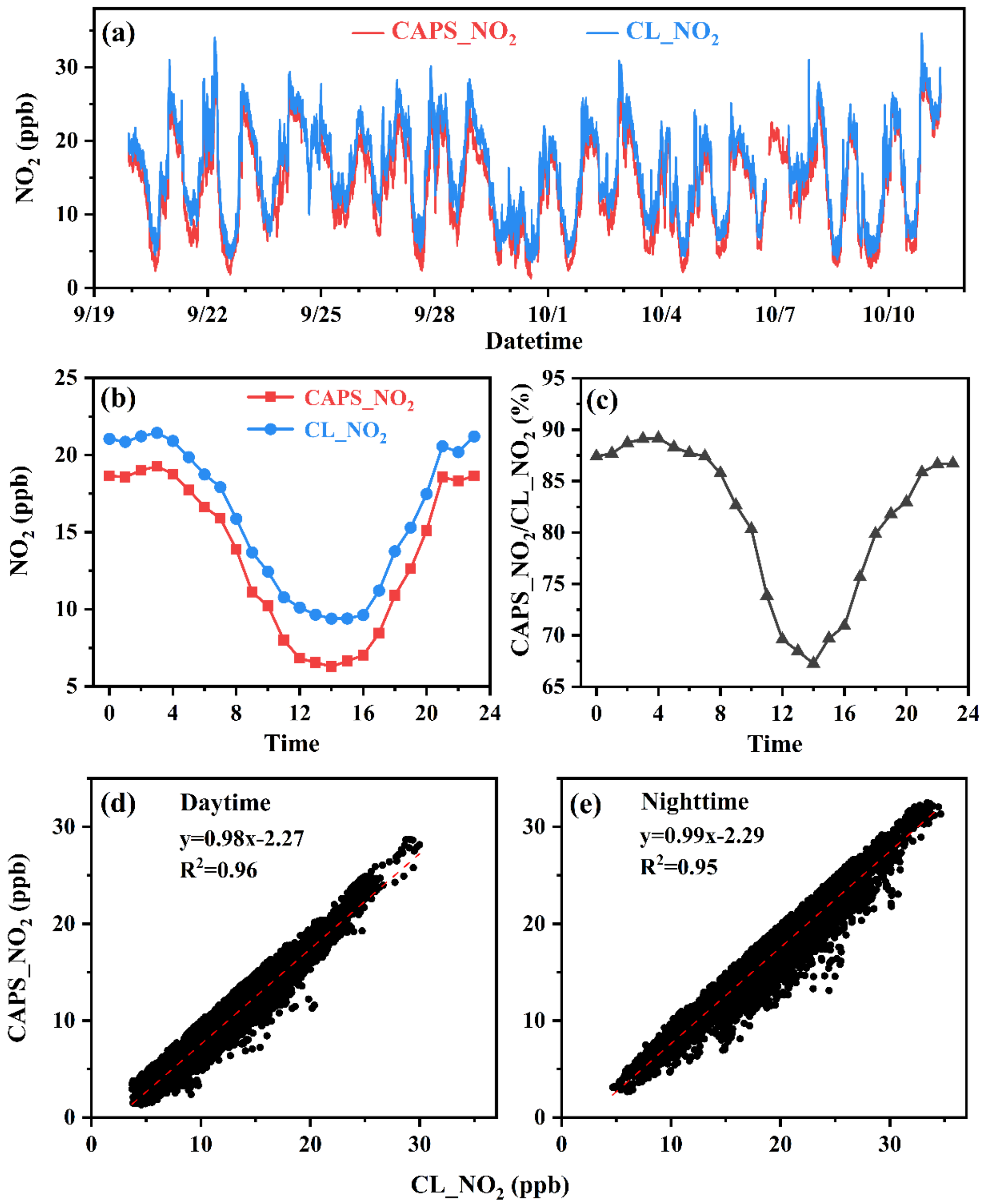

Figure S1 Time series (a) and diurnal variations (b) of CAPS_NO₂ and CL_NO₂, the diurnal variations of the fraction of CAPS_NO₂ in CL_NO₂ (c), and scatter plots with linear fits of CAPS_NO₂ versus CL_NO₂ during daytime (d) and nighttime (e).

Additionally, we sincerely appreciate the recommended reference, which provided valuable guidance for the NO_2 correction in this study. We added the relevant information and cited the suggested reference in the revised manuscript (Lines 120–122):

"The chemiluminescence (CL) technique could overestimate NO_2 concentrations due to interference from NOy (Villena et al., 2012; Wu et al., 2022a). Details of NO_2 correction were provided in Text S1 of the Supporting Information."

3. L123-124, delete the Wolfe et al before the bracket. It is the same for other similar references, such as Yan et al. (Yan et al., 2015), Zhang et al. (Zhang et al., 2019b), etc.

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Deleted the Wolfe et al., Yan et al., and Zhang et al. before the bracket. Lines 130-131 and lines 318-321 in the revised manuscript:

"For more details on this part of the model, refer to (Wolfe et al., 2016)."

"For example, (Yan et al., 2015) and (Zhang et al., 2019b) reported that during haze pollution events in Beijing in the mid-2010s, the average $PM_{2.5}$ concentration could reach approximately 130 μg m^{-3}, with levels during severe haze episodes approaching 311 μg m^{-3}."

4. Figure 1 caption, it is better to define the meaning of DHP, PEP, and CLP. The meaning of the color bar in the 2nd subfigure should also be clarified.

Response: Thanks for your valuable comments. Revision was made as the referee suggested. In the caption of Figure 1, the meanings of DHP, PEP, and CLP are clearly defined, and the meaning of the color bar the 2nd subfigure is also specified. Additionally, "WD (°)" is also labeled in the 2nd subfigure. Lines 151-157 in the revised manuscript:

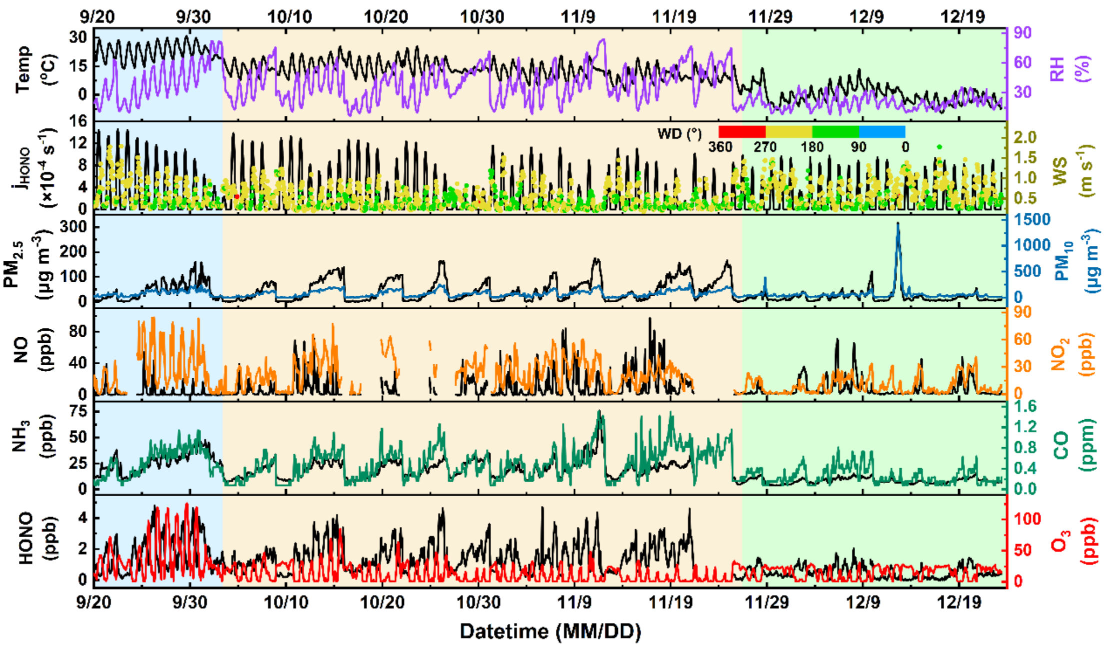

Figure 1: Hourly time series of meteorological parameters (Temp, RH, WS, j_{HONO}) and chemical species (HONO, O_3, NO, NO_2, NH_3, CO, $PM_{2.5}$, PM_{10}) concentrations from 20 September to 23 December 2022. The blue, yellow and green shades represent DHP, PEP and CLP, respectively. (DHP: Double-High Pollution Period, characterized by double-high levels of both O_3 and $PM_{2.5}$; PEP: $PM_{2.5}$ Episodic-cycle Pollution Period,

characterized by periodic cycle of $PM_{2.5}$ pollution; CLP: Clean Low Pollution Period, characterized by relatively low pollutant concentrations.) The color bar in the second subfigure represents wind direction (WD) in degrees.

5. L163-169, can you shortly explain the reasons why the pollutants concentrations are so low?

Response: Thanks for your valuable comments. During the CLP period (November 26 to December 23), pollutants concentrations were significantly lower, mainly due to a substantial reduction in anthropogenic activities. In urban Beijing, anthropogenic emissions are dominated by vehicle emissions. During the CLP period, the number of vehicles on the traffic arteries near the observation site decreased markedly, and traffic reports also showed a significant decline in the Traffic Performance Index (TPI), which indicates that the reduction in anthropogenic activities was the primary reason for the decrease in pollutants concentrations. The reasons for the reduced pollutants concentrations during the CLP period were also explained in detail in lines 258–264 of the manuscript.

6. The value used in Table 2 should be listed.

Response: Thanks for your valuable comments. The value used in Table 2 was listed as the referee suggested. Table 2 in the revised manuscript:

Source/ Loss	RACM Mechanisms	Parametrization
S_{emis}	Direct emission	EF_{emis}=0.0051
S_{NO+OH}	$NO + OH \rightarrow HONO$	k_{OH+NO}
$S_{NO_2\_g}$	$2NO_2 + H_2O \xrightarrow{\text{ground surface}} HONO + HNO_3$	$k_{het\text{-}g}=\frac{1}{8}\times v_{NO_2}\times\frac{1}{MLH}\times\gamma_g$
$S_{NO_2\_a}$	$2NO_2 + H_2O \xrightarrow{\text{aerosol surface}} HONO + HNO_3$	$k_{het\text{-}a}=\frac{1}{8}\times v_{NO_2}\times SA\times\gamma_a$
$S_{NO_2\_g,\,hv}$	$2NO_2 + H_2O + hv \xrightarrow{\text{ground surface}} HONO + HNO_3$	$k_{het\text{-}g,hv}=\frac{1}{4}\times v_{NO_2}\times\frac{1}{MLH}\times\gamma_{g,hv}\times\frac{j_{NO2}}{0.005s^{-1}}$
$S_{NO_2\_a,\,hv}$	$2NO_2 + H_2O + hv \xrightarrow{\text{aerosol surface}} HONO + HNO_3$	$k_{het\text{-}a,hv}=\frac{1}{4}\times v_{NO_2}\times SA\times\gamma_{a,hv}\times\frac{j_{NO2}}{0.005s^{-1}}$
$S_{pNO_3,\,hv}$	$pNO_3 + hv \rightarrow 0.67HONO + 0.33NO_x$	$k_{pNO_3,\,hv}=EF\times j_{HNO_3}$
L_{photo}	$HONO + hv \rightarrow OH + NO$	j_{HONO}
$L_{HONO+OH}$	$HONO + OH \rightarrow NO_2 + H_2O$	$k_{OH+HONO}$
L_{dep}	HONO deposition	$k=\frac{v_{HONO}}{BLH}$

As shown in Text S5 of the Supporting Information, the values of γ_g and $\gamma_{g,hv}$ were set to 2.94×10^{-6}, while the values of γ_a and $\gamma_{a,hv}$ were set to 3.12×10^{-5}. MLH was taken as 50 m in this observation to assess the ground-level sources of HONO (Lee et al., 2016; Xue et al., 2020; Xue et al., 2022). The enhancement factor (EF) was set to 30, a value commonly used in field observations conducted in autumn in Beijing (Zhang et al., 2022a; Xuan et al., 2024). The average dry deposition velocity of HONO (v_{HONO}) was taken as 2 cm s^{-1} (Harrison et al., 1996). k_{NO+OH}, $k_{OH+HONO}$, and j_{HNO3} were calculated in the RACM mechanisms. BLH represents boundary layer height, with units in meters (m).

7. L382-385, did you find any relationships between HONO concentrations and solar radiation? If you have, please show the data. Please refer to the publication: Explainable Machine Learning Reveals the Unknown Sources of Atmospheric HONO during COVID-19, ACS EST Air 2024, 1, 1252−1261.

Response: Thank you for your valuable comment and for recommending the reference. We analyzed the relationships between $HONO_{unknown}$, $P_{unknown}$, and solar radiation. Hourly solar radiation data were derived from the ERA5 reanalysis dataset (ECMWF). As shown in the revised Figure R1 (see below), $HONO_{unknown}$ exhibited a weak positive correlation with solar radiation (R^2 = 0.23), while $P_{unknown}$ showed a moderate positive correlation (R^2 = 0.36). These results indicated that both $HONO_{unknown}$ and $P_{unknown}$ were influenced by solar radiation. However, the degree of correlation differs. $P_{unknown}$ represented the production rate of HONO and was directly driven by photochemical processes during daytime, which were enhanced with increasing solar radiation. In contrast, $HONO_{unknown}$ concentrations were determined by a balance between production and removal processes (e.g., photolysis reaction, heterogeneous reaction and homogeneous reaction), which weakened its direct correlation with solar radiation. Therefore, the relatively stronger correlation observed for $P_{unknown}$ supported the hypothesis that unknown HONO sources were photochemically driven during daytime, while the weaker correlation of $HONO_{unknown}$ was consistent with its influence by multiple processes.

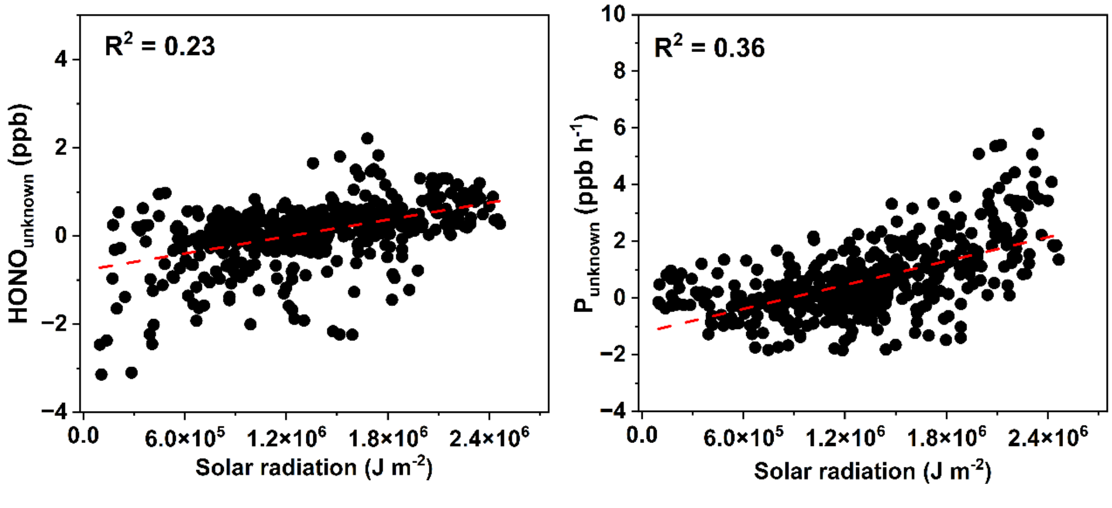

Figure R1. Correlations between $HONO_{unknown}$ and $P_{unknown}$ with solar radiation. The red dashed lines represent linear regression fits.

In addition, we carefully considered your advice and cited the recommended reference (Explainable Machine Learning Reveals the Unknown Sources of Atmospheric HONO during COVID-19, ACS EST Air, 2024, 1, 1252–1261) in the revised manuscript. Lines 411-413 in the revised manuscript:

"First, pathways related to solar radiation and OH radicals should be considered in future studies on daytime HONO sources, as also suggested by recent findings using explainable machine learning approaches (Gao et al., 2024)."

8. The implications of the research should be clarified. Such as in L419-433, the results may indicate that control vehicle emissions could be an effective measures to reduce air pollution, while more measures should be integrated during the haze periods.

Response: Thanks for your valuable comments. The implications of the research were clarified as the referee suggested. Lines 467-471 in the revised manuscript:

"These results had important policy implications for air pollution control. The study indicated that controlling vehicle emissions might be an effective measure to reduce HONO concentrations and improve air quality. However, during haze pollution periods, it is necessary to complement vehicle emission control with integrated multi-source measures, such as reducing NO_2 and NH_3 emissions, to limit the secondary formation of HONO and thereby more effectively reduce air pollution."

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A point-by-point response to Referee #2

We sincerely appreciate Referee #2 for the valuable comments and constructive suggestions that help improve the quality of our manuscript. The following is a point-by-point response to address the referee's comments. The original comments are shown in *black*, and our corresponding responses are presented in *blue*. The new or modified contents in the revised manuscript are marked in *red*.

Comments from Referee #2:

The manuscript uses measurements of gas-phase species and aerosols together with meteorological parameters to separate the measurements into three distinct periods. For each period they use a model to evaluate the sources of HONO and how they vary between the three phases due to changes in anthropogenic emissions. My major concern with the study is the use of a NOx analyzer (Thermo Fisher 42i), which according to the manual uses a molybdenum converter for the NO_2 measurements. This is problematic since molybdenum converters are known to overestimate NO_2 due to conversion of PANs and other nitrogen containing compounds and NO_2 is a key component of the analysis. Additionally, some clarification is required throughout the manuscript, which is commented as minor comments or technical comments to improve the readability of the manuscript.

I recommend the manuscript be published when these concerns are addressed.

Response: Many thanks to Referee #2 for the thorough review and valuable comments on our manuscript. We fully understand your major concern regarding the potential overestimation of NO_2 when using the NOx analyzer (Thermo Fisher 42i), which is indeed important for the reliability of our study. In the revised manuscript, we will provide a detailed clarification and add corresponding comparison results and analyses. Meanwhile, we will carefully address and revise general comments, major comments, minor comments, and technical comments to further improve the readability and scientific quality of the manuscript. The following are point-by-point responses to the referee's comments.

General comments:

I would suggest changing "anthropogenic activities variations" to "variations in anthropogenic activities" throughout the paper as it is easier to read.

Response: Thanks for your helpful suggestions. We agree that "variations in anthropogenic activities" is clearer and easier to read. We revised the wording accordingly throughout the manuscript.

Major comments:

Line 114-115: Since the NOx analyzer uses a molybdenum converter, it also converts organic nitrates into NO_2/NO and potentially also particulate nitrates. Do you somehow take that into account when using the NO_2 measurements? How often is the sensitivity of the different channels calibrated? If it isn't taken into account, can you estimate an uncertainty on the measurements?

Response: Thanks for your valuable comments. Revision was made as the referee suggested. We corrected CL_NO_2 (Thermo Scientific, Model 42i NOx analyzer) using interference-free CAPS_NO_2 measurements (Teledyne API-N500 NOx analyzer), and provided rigorous field comparison evidence. To improve the precision and accuracy of the NO_2 correction, we established separate daytime and nighttime linear regressions. All relevant parameters were recalculated with the corrected NO_2, and the model simulations were rerun. The correction methodology and its impacts were described in detail in the Supporting Information (see Text S1 and Figure S1).

Text S1 in the Supporting Information:

"As the most important precursor of HONO, accurate measurement of NO_2 was crucial for analyzing HONO formation. A commercial Thermo Scientific analyzer (42i) used in this study could specifically detect NO. The measurement of NO_2 was achieved by converting NO_2 to NO through a molybdenum converter. However, the chemiluminescence (CL) technique could overestimate NO_2 concentrations because of the interference of NOy. These interferences included HONO, HNO_3, HNO_4, N_2O_5, NO_3, peroxyacetyl nitrate (PANs, $RC(O)OONO_2$), organic nitrates ($RONO_2$), and peroxynitrates ($ROONO_2$) (Villena et al., 2012; Wu et al., 2022). Therefore, the NO_2 measured by the CL-NOx analyzer represented the sum of real NO_2 and these interfering species. In contrast, the commercial Teledyne API-N500 NOx analyzer was based on cavity attenuated phase shift (CAPS) technique. It could provide direct absorption measurement of NO_2 at 450 nm in the blue region of the electromagnetic spectrum, allowed fast and accurate detection of NO_2 without interference from water vapor. The only known potential interferences in the typical ambient environment were dicarbonyl compounds such as glyoxal and methylglyoxal, whose concentrations were usually much lower than NO_2 mixing ratios (Kebabian et al., 2008). Therefore, NO_2 measured by the CAPS-NOx analyzer (CAPS_NO_2) could be used to correct the NO_2 measured by the CL-NOx analyzer (CL_NO_2).

We conducted a NO_2 field campaign at the ICCAS site from September 19 to October 11, 2023, to compare the performance of the CL-NOx and CAPS-NOx analyzers. The sampling inlets of both instruments were placed at the same location, with identical sampling tube lengths, and the analyzers were housed in the same indoor environment to minimize external interference. The results showed that CAPS_NO_2 and CL_NO_2 exhibited similar temporal variations (Figure S1(a) and S1(b)). Notably, CL_NO_2 was consistently higher than CAPS_NO_2, with a more pronounced difference during the daytime. This discrepancy was mainly attributed to elevated NOy concentrations caused by enhanced photochemical reactions. Consequently, the fraction of CAPS_NO_2 in CL_NO_2 displayed a distinct diurnal pattern, being higher at night and lower during the day (Figure S1(c)), which was consistent with previous findings (Xue et al., 2022; Zhang et al., 2022c). Based on this result, we applied separate calibrations for daytime (07:00-18:00 LT) and nighttime (19:00-next 06:00 LT) data. The results indicated strong linear correlations between CAPS_NO_2 and CL_NO_2 during both periods ($R^2 = 0.96$ for daytime and $R^2 = 0.95$ for nighttime). The regression equations were “y = 0.98x − 2.27” for daytime and “y = 0.99x − 2.29” for nighttime, where y represented CAPS_NO_2 and x represented CL_NO_2 (Figure S1(d) and S1(e)). Using these relationships to correct the NO_2 data obtained in this study provided a more reasonable estimation of true NO_2 concentrations and offered a reliable basis for further analysis.”

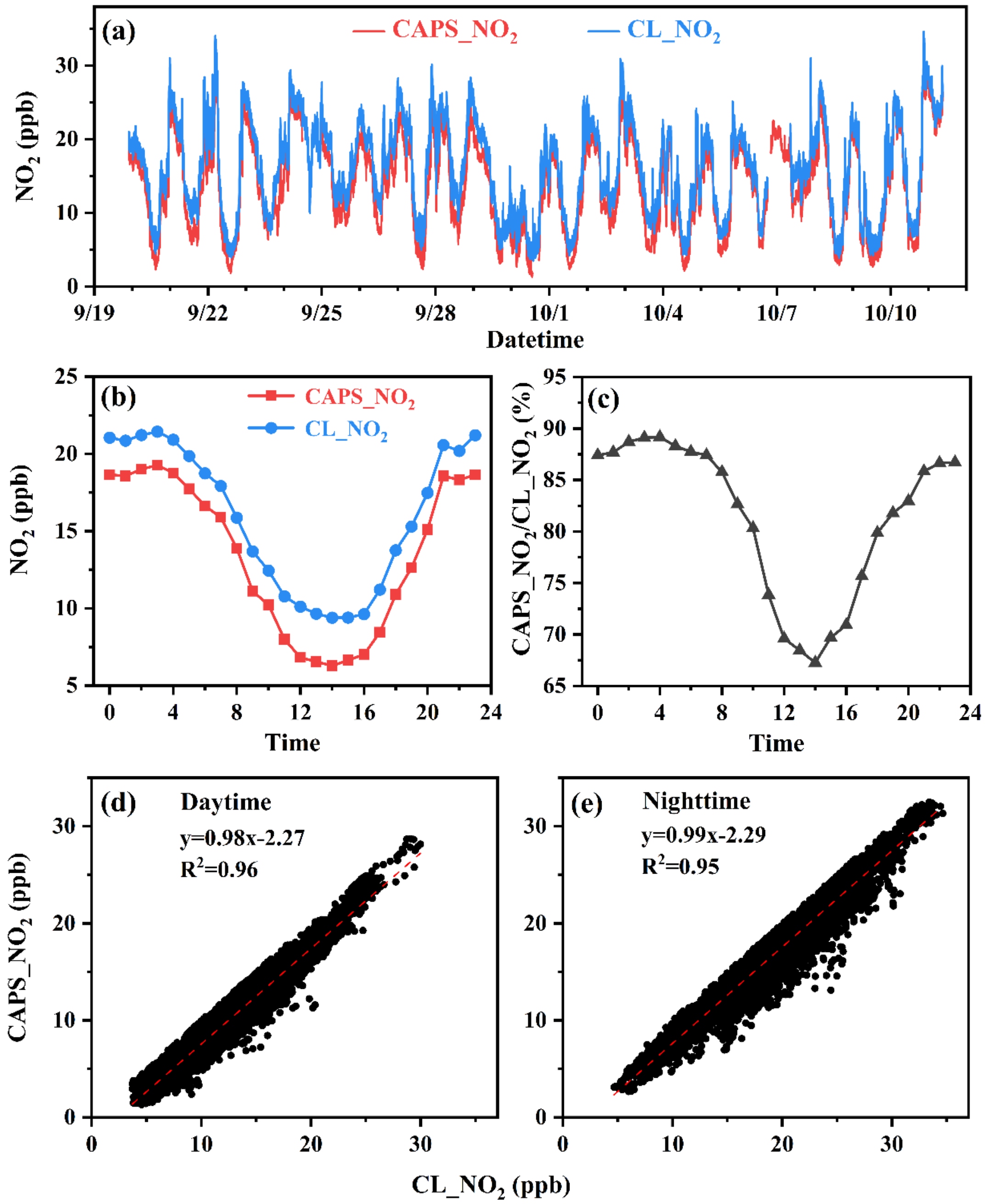

Figure S1 Time series (a) and diurnal variations (b) of CAPS_NO_2 and CL_NO_2, the diurnal variations of the fraction of CAPS_NO_2 in CL_NO_2 (c), and scatter plots with linear fits of CAPS_NO_2 versus CL_NO_2 during daytime (d) and nighttime (e).

Additionally, we added the information in the revised manuscript (Lines 120–122):

“The chemiluminescence (CL) technique could overestimate NO_2 concentrations due to interference from NOy (Villena et al., 2012; Wu et al., 2022a). Details of NO_2

correction were provided in Text S1 of the Supporting Information."

Minor comments:

Line 40: Something seems to be missing in the following sentence: "The more severe pollution, and the higher contribution of HONO to primary OH radicals (70–92 %)."

Response: Thanks for your helpful suggestions. Revision was made as the referee suggested. Lines 42–43 in the revised manuscript:

"During more severe pollution, the contribution of HONO to primary OH radical was higher (70–92%)."

Line 56-63: The part of the paragraph between "Over the last decade" and "simultaneous control of both $PM_{2.5}$ and O_3" require some grammatical rephrasing. If the climate policies were implemented prior to another past event, then the use of past perfect tense is good, however, when writing "Over the last decade" then it should just be written in past tense. If you add when the air pollution control focus switched in line 62, then that becomes the other event in the past.

Response: Thanks to the referee for the clear explanation regarding the use of tense. We agree that the phrase "Over the last decade" should be followed by past tense rather than past perfect tense, unless contrasted with another event in the past. Accordingly, we revised the paragraph in Lines 59–66 to use the past tense consistently.

"Over the past decade, Beijing implemented various measures, including the Clean Air Action Plan in 2013 and the Three-Year Action Plan from 2018 to 2020, and moved many heavy-polluting industries out of Beijing to control industrial pollution (Zhang et al., 2016; Chan and Yao, 2008). Additionally, the control of vehicle emissions and coal combustion in Beijing was one of the key tasks (Zhang et al., 2016). With the implementation of these policies, $PM_{2.5}$ concentration decreased rapidly, while O_3 concentration increased year by year in Beijing. Moreover, despite the reduction in nitrogen oxides (NOx) emissions, the particulate nitrate (pNO_3) concentration and its proportion in $PM_{2.5}$ increased (Zong et al., 2022). The air pollution control focus shifted from single $PM_{2.5}$ control to the simultaneous control of both $PM_{2.5}$ and O_3 (Liu et al., 2020; Ye et al., 2023)."

Line 61: In the sentence "the nitrate (NO_3^-) concentration and its proportion in $PM_{2.5}$ had increased", what do you mean by nitrate? Is it particulate nitrate, organic nitrates, inorganic nitrates, nitrate radicals? I would suggest defining it the first time you use it, since it is used throughout the manuscript.

Response: Thanks to the referee for this helpful comment, and we apologize for the lack of clarity in our original wording. Here "nitrate" refers to particulate nitrate (pNO_3).

To avoid confusion, we clarified this definition when it first appeared in the manuscript and used pNO_3 consistently throughout the manuscript to denote particulate nitrate, as suggested in the second of the "technical comments" given by the referee. Lines 63–65 in the revised manuscript:

"Moreover, despite the reduction in nitrogen oxides (NOx) emissions, the particulate nitrate (pNO_3) concentration and its proportion in $PM_{2.5}$ had increased (Zong et al., 2022)."

Line 136-137: You write "As shown in Table 1, the highest HONO concentration in this study was generally higher than in other studies.", however, 30% of the previous studies have higher maximum HONO concentrations than your study according to Table 1, so maybe rephrase it to represent that.

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 143-145 in the revised manuscript:

"As shown in Table 1, the highest HONO concentration in this study was comparable to or higher than in several previous studies, though still lower than the highest values reported by Gu et al. (2022)."

Figure 1: Please define DHP, PEP, and CLP here since the figure is described before the definitions. And describe the colourbar.

Response: Thanks for your valuable comments. Revision was made as the referee suggested. In the caption of Figure 1, the meanings of DHP, PEP, and CLP are clearly defined, and the meaning of the color bar the 2^{nd} subfigure is also specified. Additionally, "WD (°)" is also labeled in the 2^{nd} subfigure. Lines 151-157 in the revised manuscript:

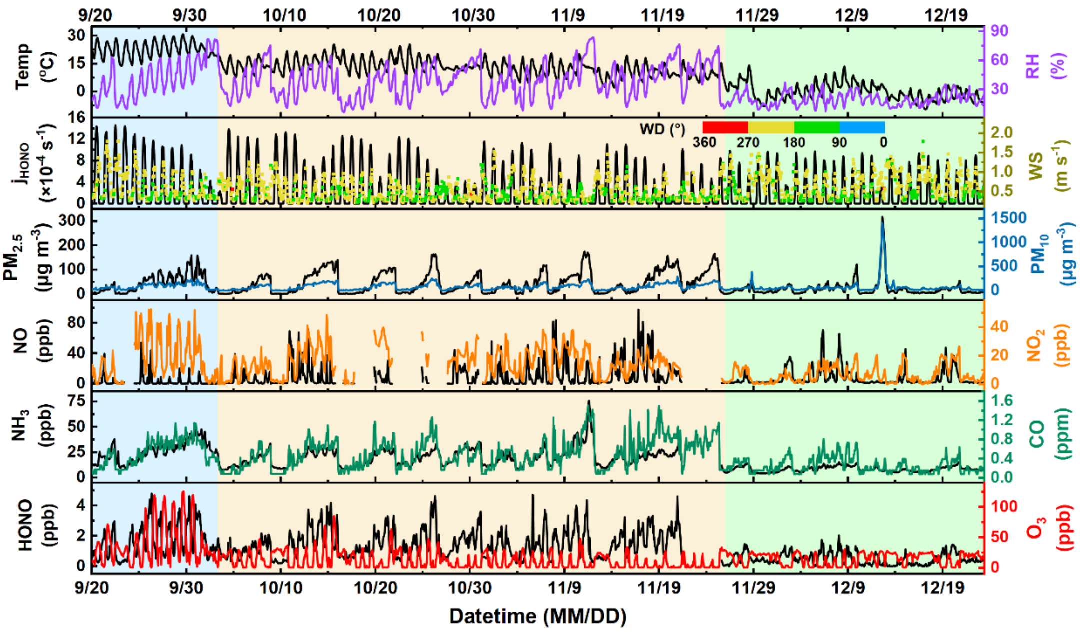

Figure 1: Hourly time series of meteorological parameters (Temp, RH, WS, j_{HONO}) and chemical species (HONO, O_3, NO, NO_2, NH_3, CO, $PM_{2.5}$, PM_{10}) concentrations from 20 September to 23 December 2022. The blue, yellow and green shades represent DHP, PEP and CLP, respectively. (DHP: Double-High Pollution Period, characterized by double-high levels of both O_3 and $PM_{2.5}$; PEP: $PM_{2.5}$ Episodic-cycle Pollution Period, characterized by periodic cycle of $PM_{2.5}$ pollution; CLP: Clean Low Pollution Period, characterized by relatively low pollutant concentrations.) The color bar in the second subfigure represents wind direction (WD) in degrees.

Line 195: When you mention NO_3^- formation, do you mean particulate nitrate? Because if it is particulate nitrate, do you then mean that particulate nitrate formation leads to $PM_{2.5}$ pollution or does this part of the sentence only refer to the low WS and BLH?

Response: Thanks to the referee for pointing this out, and we apologize for the lack of clarity in our original wording. Here "NO_3^-" indeed refers to particulate nitrate (pNO_3). We intended to state that relatively high NO_2 concentrations and high RH promoted the formation of pNO_3, which is an important component of $PM_{2.5}$. Meanwhile, low WS resulted in weaker diffusion, further contributing to the recurrence of $PM_{2.5}$ pollution. Lines 208-211 in the revised manuscript:

"However, relatively high NO_2 concentrations and high RH promoted the formation of pNO_3 (Xu et al., 2019), which was an important component of $PM_{2.5}$. Meanwhile, low WS resulted in weaker diffusion, further contributing to the recurrence of $PM_{2.5}$ pollution (Liu et al., 2023)."

Line 201-203: You write "During the DHP and PEP, NO, NO_2, CO and NH_3 showed significant peaks during the morning rush hour (7:00–8:00 LT) due to vehicle emissions, then remained at lower levels throughout the daytime until concentrations began to rise again during the evening rush hour and built up during the night.”. This seems like a overgeneralisation since both NO_2 and NH_3 only show small if any enhancement during the morning rush hour in the PEP phase, CO doesn’t reach lower levels after the increase during DHP and as you write in the following sentence NO doesn’t increase at nighttime during DHP.

Response: Thanks for your valuable comments, and we apologize for the lack of clarity in our original wording. We agree that our original wording in Lines 201–203 was too general and did not fully capture the differences among species and periods. In the revised manuscript, we rephrased the description to more accurately reflect the observed behaviors. Specifically, compared with the CLP, NO_2 indeed exhibited a more obvious morning peak during the PEP, while NH_3 showed only a weaker enhancement. CO did not decrease after the morning peak during the DHP. This is because $PM_{2.5}$ concentrations remained high from 09:00–13:00 LT, and given the strong correlation between CO and $PM_{2.5}$, CO concentrations also remained elevated, only starting to decrease after 13:00 LT. In addition, NO did not increase at night during DHP due to the stronger O_3 consumption of NO, as explained later in the text. To address this, we revised the sentence to avoid overgeneralization and to provide a more accurate description of the variations across species and phases. Lines 215-221 in the revised manuscript:

“During the DHP and PEP, traffic-related peaks of NO, NO_2, CO and NH_3 were generally observed during the morning rush hour (7:00–8:00 LT), then generally remained at lower levels throughout the daytime until concentrations began to rise again during the evening rush hour and built up during the night. The magnitude and subsequent variations differed by species and periods. For example, NO_2 and NH_3 showed only small enhancements in the PEP, and CO remained elevated after the morning peak during DHP and decrease after 13:00 LT. Notably, the nighttime NO concentration was also very low during the DHP, staying between 0.46–1.17 ppb, which was due to the stronger O_3 consumption of NO (Kurtenbach et al., 2012).”

Line 205: NO does not look lower during CLP.

Response: Thanks to the referee for pointing this out, and we apologize for the lack of clarity in our original wording. We agree that the statement in the original text was too general. In fact, the decrease of NO during CLP depends on which period it is compared with. When compared with the PEP, which was also characterized by lower O_3 levels, NO concentrations during CLP were indeed lower, reflecting reduced vehicle emissions. However, compared with the DHP, where O_3 concentrations were relatively high and NO was already suppressed, the NO level during CLP was not significantly lower and even appeared slightly higher. To avoid confusion, we revised the text to more accurately describe these differences. Line 222 in the revised manuscript:

“During the CLP, the concentrations of NO_2, CO and NH_3 were lower, and their diurnal variations were not obvious."

Line 209: What do you mean by vertical contrast and horizontal comparison?

Response: Thanks to the referee for pointing this out, and we apologize for the lack of clarity in our original wording. By "vertical contrast", we intended to describe the distinct differences in pollution characteristics among the three periods (DHP, PEP, and CLP), that is, the distinct contrast formed among these three periods. By "horizontal comparison", we meant the comparison of our results with those from the same periods reported in other studies. Lines 224-226 in the revised manuscript:

"The pollution characteristics of the three periods (DHP, PEP, and CLP) exhibited both distinct differences and certain similarities, forming a sharp vertical contrast while also allowing for a horizontal comparison, meaning cross-study comparisons with published results for the same periods."

Line 269-270: You write "exhibited significant increases in the evening (~19:00 LT) and early morning (~6:00 LT) during the DHP and PEP", but in Figure 3a it looks like DHP is continuously increasing over the night and PEP is fairly flat.

Response: Thanks to the referee for this valuable comment, and we sincerely apologize for the lack of clarity in our original wording. We agree that in Figure 3a, $HONO_{emis}$ shows a continuous increase throughout the night during DHP, while $HONO_{emis}$ appears relatively flat during PEP. Our original statement in Lines 269–270 was not intended to describe the entire nighttime trend, but rather to highlight the specific periods of 19:00–20:00 and 05:00–06:00 LT, which correspond to the evening and morning rush hours and best represent enhanced vehicle emissions. To avoid misunderstanding, we revised the sentence to clarify this focus and to make the comparison with CLP more precise. Lines 288-291 in the revised manuscript:

"$HONO_{emis}$ exhibited significant increases during the evening (19:00–20:00 LT) and early morning (05:00–06:00 LT) rush hours in both DHP and PEP, reflecting stronger vehicle emissions compared with CLP."

Line 305-307: Maybe add that the enhanced oxidation of organic and inorganics during DHP is consistent with the high O_3 concentrations observed.

Response: Thanks to the referee for this valuable comment. We agree that the enhanced oxidation of organic and inorganic components during DHP is consistent with the high O_3 concentrations observed. To reflect this, we revised the sentence. Lines 327-330 in the revised manuscript:

"Notably, $HONO_{corr}$ exhibited the strongest correlation with both NO_2 and $NO_2 \times PM_{2.5}$ during the DHP, likely due to enhanced oxidation of organic and inorganic components, consistent with the high O_3 concentrations observed, which altered the surface reactivity and consequently promoted NO_2 conversion to HONO (George et al., 2015; Ndour et al., 2008)."

Table 2: The references used for the parameterization should be mentioned.

Response: Thanks for your valuable comments. The references used for the parameterization in Table 2 was listed as the referee suggested. Table 2 in the revised manuscript:

Source/ Loss	RACM Mechanisms	Parametrization
S_{emis}	Direct emission	EF_{emis}=0.0051
S_{NO+OH}	$NO + OH \rightarrow HONO$	k_{OH+NO}
$S_{NO_2\_g}$	$2NO_2 + H_2O \xrightarrow{\text{ground surface}} HONO + HNO_3$	$k_{het\text{-}g}=\frac{1}{8}\times v_{NO_2}\times\frac{1}{MLH}\times\gamma_g$
$S_{NO_2\_a}$	$2NO_2 + H_2O \xrightarrow{\text{aerosol surface}} HONO + HNO_3$	$k_{het\text{-}a}=\frac{1}{8}\times v_{NO_2}\times SA\times\gamma_a$
$S_{NO_2\_g,\,hv}$	$2NO_2 + H_2O + hv \xrightarrow{\text{ground surface}} HONO + HNO_3$	$k_{het\text{-}g,hv}=\frac{1}{4}\times v_{NO_2}\times\frac{1}{MLH}\times\gamma_{g,hv}\times\frac{j_{NO2}}{0.005s^{-1}}$
$S_{NO_2\_a,\,hv}$	$2NO_2 + H_2O + hv \xrightarrow{\text{aerosol surface}} HONO + HNO_3$	$k_{het\text{-}a,hv}=\frac{1}{4}\times v_{NO_2}\times SA\times\gamma_{a,hv}\times\frac{j_{NO2}}{0.005s^{-1}}$
$S_{pNO_3,\,hv}$	$pNO_3 + hv \rightarrow 0.67HONO + 0.33NO_x$	$k_{pNO_3,\,hv}=EF\times j_{HNO_3}$
L_{photo}	$HONO + hv \rightarrow OH + NO$	j_{HONO}
$L_{HONO+OH}$	$HONO + OH \rightarrow NO_2 + H_2O$	$k_{OH+HONO}$
L_{dep}	HONO deposition	$k=\frac{v_{HONO}}{BLH}$

As shown in Text S5 of the Supporting Information, the values of γ_g and $\gamma_{g,hv}$ were set to 2.94×10^{-6}, while the values of γ_a and $\gamma_{a,hv}$ were set to 3.12×10^{-5}. MLH was taken as 50 m in this observation to assess the ground-level sources of HONO (Lee et al., 2016; Xue et al., 2020; Xue et al., 2022). The enhancement factor (EF) was set to 30, a value commonly used in field observations conducted in autumn in Beijing (Zhang et al., 2022a; Xuan et al., 2024). The average dry deposition velocity of HONO (v_{HONO}) was taken as 2 cm s^{-1} (Harrison et al., 1996). k_{NO+OH}, $k_{OH+HONO}$, and j_{HNO3} were calculated in the RACM mechanisms. BLH represents boundary layer height, with units in meters (m).

Line 340-342: You write in the SI that you use EF=30 for the photolysis of particulate nitrate, however, studies have reported values between 1 and 700 for aerosols (Ye et al., 2016, Romer et al., 2018, Ye et al., 2017) and up to 1700 for urban grime (Baergen and Donaldson, 2013). Recent studies have found that the enhancement factor (EF) for photolysis of particulate nitrate depends on different aerosol parameters and for example decrease with increasing particulate nitrate (Andersen et al., 2023, Sommariva et al., 2023, Rowlinson et al., 2025). These dependencies are not incorporated in your model and would maybe give a different effect than what you observed (increasing importance of photolysis of particulate nitrate to the HONO formation with increasing particulate nitrate). While it is probably outside the scope of this paper to investigate the impact of different parameterizations of the EF, it would be good with a couple of sentences to discuss these effects and how it might impact your results.

C. Ye et al., Rapid cycling of reactive nitrogen in the marine boundary layer. Nature**532**, 489–491 (2016).

C. Ye et al., Photolysis of particulate nitrate as a source of HONO and NO_x. Environ. Sci. Technol.**51**, 6849–6856 (2017)

P. S. Romer et al., Constraints on aerosol nitrate photolysis as a potential source of HONO and NO_x. Environ. Sci. Technol.**52**, 13738–13746 (2018).

A. M. Baergen, D. J. Donaldson, Photochemical renoxification of nitric acid on real urban grime. Environ. Sci. Technol.**47**, 815–820 (2013).

S. T. Andersen et al., Extensive field evidence for the release of HONO from the photolysis of nitrate aerosols.Sci. Adv.**9**, eadd6266(2023)

R. Sommariva et al., Factors Influencing the Formation of Nitrous Acid from Photolysis of Particulate Nitrate. JPCA 127, 9302-9310 (2023)

M. J. Rowlinson et al., Observations of tropospheric HONO are incompatible with understanding of atmospheric chemistry, EGUsphere [preprint] (2025)

Response: Thanks to the referee for this valuable comment. As you correctly pointed out, the EF for pNO_3 photolysis varies widely across different studies and is influenced by various aerosol parameters. Therefore, we added a sensitivity analysis in the Supporting Information to evaluate the impact of EF uncertainty on the contribution of pNO_3 photolysis to HONO formation. Text S6 and Table S3 in the Supporting Information:

"EF represented the enhancement factor of the photolysis rate of pNO_3 relative to that of HNO_3. Laboratory studies reported EF values between 1 and 700 for aerosols (Romer et al., 2018; Ye et al., 2016; Ye et al., 2017), and experimental values up to 1700 for urban grime (Baergen and Donaldson, 2013). However, EF is widely considered to carry substantial uncertainty, which can translate into uncertainty in HONO concentrations. In this study, we adopted a moderate EF (=30) commonly used for autumn in Beijing (Zhang et al., 2022a; Xuan et al., 2024). In addition, to comprehensively evaluate the potential impact of EF uncertainty on the results, a sensitivity analysis was conducted by decreasing and increasing the EF by one order of magnitude (i.e., EF=3 and EF=300). The corresponding changes in HONO concentrations during the three periods were summarized in Table S3. When EF=3, the changes were approximately 3.2 %, 3.4 %, and 2.1 % during DHP, PEP, and CLP, respectively, indicating that the variation in the contribution of pNO_3 photolysis to

HONO formation was negligible compared with that under EF = 30. In contrast, when EF=300, the changes were 31.5 %, 34.1 %, and 20.5 %, respectively, suggesting that the contribution of pNO_3 photolysis to HONO formation increased slightly relative to the EF=30. These results demonstrated that the EF value could influence the contribution of pNO_3 photolysis to HONO formation, highlighting the importance of EF in quantitatively constraining the HONO budget."

Table S3 Sensitivity study with EF uncertainty for HONO formation processes.

EF	DHP	PEP	CLP
3	-3.2 %	-3.4 %	-2.1%
300	31.5 %	34.1 %	20.5 %

Additionally, we added the information in the revised manuscript (Lines 367–370):

"A sensitivity analysis (Text S6 and Table S3) showed that variations in the EF had a limited effect on HONO formation when EF = 3, but led to a noticeable increase when EF = 300, indicating that EF could influence the contribution of pNO_3 photolysis to HONO production. These results highlight the importance of EF in quantitatively constraining the HONO budget."

Line 424-425: You write "NO_3^- photolysis accounted for 12.6 %, 11.8 %, and 4.8 %, consistent with $PM_{2.5}$ concentrations in three periods, and indicating increasing NO_3^- fractions in $PM_{2.5}$ under more polluted conditions.", but is that really what you mean? Since the NO_3 is approximated based on the mass fraction of PM_1 in $PM_{2.5}$ (line 100 in the manuscript) and you use the same EF to determine the HONO production for NO_3^- photolysis is it not just an indication that you have significantly more aerosols available with increasing pollution?

Response: Thanks to the referee for this thoughtful comment. Our intention was to emphasize that the enhanced contribution of pNO_3 photolysis during more polluted periods was consistent with $PM_{2.5}$ concentrations. Since the pNO_3 concentration was approximated using the $PM_1/PM_{2.5}$ ratio and a constant EF was applied, this trend is more reasonably interpreted as reflecting higher $PM_{2.5}$ concentrations, rather than a compositional change. In our analysis, the same EF was applied across three periods in order to ensure consistency and comparability of the HONO source budget within this observation study. We are aware that the choice of EF is a challenging issue. Therefore, a sensitivity analysis of the EF was included in the Supporting Information (Text S6 and Table S3) to assess the potential influence of EF uncertainty on these results.

To improve clarity and avoid potential ambiguity, we revised the sentence to focus only on the quantitative contribution of pNO_3 photolysis across the three periods and its consistency with $PM_{2.5}$ concentrations. Lines 456-458 in the revised manuscript:

"pNO_3 photolysis accounted for 12.7 %, 11.7 %, and 5.0 %, consistent with $PM_{2.5}$

concentrations in the three periods."

Technical comments:

Line 19-20: Change "a comprehensive observation" to "comprehensive observations"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 19-21 in the revised manuscript:

"Therefore, we investigated the impact of variations in anthropogenic activities on HONO formation based on comprehensive observations conducted in urban Beijing during autumn and winter of 2022."

Line 25: I would suggest writing particulate nitrate as pNO_3 instead of NO_3^- to avoid people misunderstanding it for NO_3 radicals.

Response: Thanks for your valuable comments. We agree that using "pNO_3" is clearer and avoids confusion with NO_3 radicals. Following the suggestion, we revised "NO_3^-" to "pNO_3" throughout the manuscript to avoid misunderstanding.

Line 65: change "development of second pollutions in Beijing" to "development of secondary pollution in Beijing"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 67-69 in the revised manuscript:

"Previous studies have highlighted how such meteorological conditions facilitate the development of secondary pollution in Beijing, with weak southerly winds often driving pollution from industrial regions (Guo et al., 2014; Zheng et al., 2015)."

Line 71: delete "had" in "Hereby, we had conducted"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 75-77 in the revised manuscript:

"Hereby, we conducted a field observation campaign in urban Beijing from 20 September to 23 December 2022, covering the autumn and winter seasons when O_3 and $PM_{2.5}$ pollution frequently occurred."

Line 75: change "During this observations" to either "During these observations" or "During this campaign" or "During this observation period"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 78-79 in the revised manuscript:

"During this campaign, stagnant meteorological conditions predominated, with low wind speeds and southerly winds."

Line 78: replace "to" with "on" in "impact to secondary"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 80-82 in the revised manuscript:

"It provided a unique opportunity to identify HONO sources and their potential impact on secondary pollution formation in urban Beijing, which has been rarely studied in the past."

Line 78: replace "studies" with "studied"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 80-82 in the revised manuscript:

"It provided a unique opportunity to identify HONO sources and their potential impact on secondary pollution formation in urban Beijing, which has been rarely studied in the past."

Line 82: replace "provided" with "provide"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 85-87 in the revised manuscript:

"In summary, through continuous field observations and model simulations, we provide direct evidence that reducing anthropogenic activities is crucial for controlling wintertime HONO formation in Beijing, providing a direct basis for formulating effective air pollution control strategies."

Line 123: remove "(Wolfe et al., 2016)" in "refer to Wolfe et al (Wolfe et al., 2016)"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 130-131 in the revised manuscript:

"For more details on this part of the model, refer to (Wolfe et al., 2016)."

Line 129: replace "illustrated" with "illustrates"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 136-137 in the revised manuscript:

"Figure 1 illustrates the hourly time series of meteorological parameters and chemical species concentrations during 20 September to 23 December 2022."

Line 132: replace "observation" with "campaign" or "observation period"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 138-140 in the revised manuscript:

"Throughout the entire campaign, there was a significant variation of temperature (Temp) and relative humidity (RH) due to the span across autumn and winter."

Line 132: Add "the" to write "due to the span across"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 138-140 in the revised manuscript:

"Throughout the entire campaign, there was a significant variation of temperature (Temp) and relative humidity (RH) due to the span across autumn and winter."

Line 135: delete "the" in front of Beijing

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 141-142 in the revised manuscript:

"The meteorological conditions represented typical stagnant conditions that promoted the accumulation of pollutants in Beijing."

Line 147-148: replace "when" with "where" to write "there were 6 days O_3 pollution where the daily maximum"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 159-161 in the revised manuscript:

"According to the National Ambient Air Quality Standards (NAAQS), during the observation period, there were 6 days O_3 pollution where the daily maximum 8-hour average concentration of O_3 exceeded the Grade II of NAAQS (160 $\mu g\ m^{-3}$, equivalent to 82 ppb at 25°C and 1013.25 hPa)."

Line 151: replace "accompanying" with "accompanied"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 162-163 in the revised manuscript:

"These days with O_3 pollution were all concentrated between September 20 and October 2, accompanied by high $PM_{2.5}$ concentrations (up to 150 $\mu g\ m^{-3}$)."

Line 181: replace "parameter" with "parameters"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 194-195 in the revised manuscript:

"Detailed diurnal variations of meteorological parameters are provided in Text S3 and Figures S2–S3 in the Supporting Information."

Line 200: replace "gases" with "gas" and add "the" to write "between the three periods"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 214-215 in the revised manuscript:

"For the diurnal variations of gas pollutants, there were significant differences between the three periods."

Figure 2 and 3 text: replace "line graphs" with "lines"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 229-230 and lines 299-300 in the revised manuscript:

"Figure 2: The diurnal variations of chemical species (HONO, NO, NO_2, NH_3, CO, O_3 $PM_{2.5}$) and meteorological parameters (Temp, RH) during three periods. The blue, red, and black dotted lines represent DHP, PEP and CLP, respectively."

"Figure 3: The hourly variations of (a) $HONO_{emis}$ and (b) $HONO_{emis}$/HONO at nighttime during three periods. The blue, red, and black dotted lines represent DHP, PEP and CLP, respectively."

Line 213 and 414: I would suggest adding "mixing ratios" after higher

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 231-232 and lines 444-445 in the revised manuscript:

"HONO and NOx (the main precursors of HONO) exhibited similar diurnal variation trends, with higher mixing ratios at night and lower during the day."

"HONO exhibited similar diurnal variation trends in the three periods, with higher mixing ratios at night and lower during the day."

Line 217, 224, 225, 226 and 227: I would suggest adding "the" in front of HONO concentration

Response: Thanks for your valuable comments. Revisions was made as the referee suggested. Lines 234-236, line 242, lines 243-244, lines 244-245 and lines 245-246 in the revised manuscript:

"Subsequently, due to the absence of photolysis reactions and the arrival of the evening rush hour, the HONO concentration began to accumulate, remaining at a high level throughout the night (1.94–2.33 ppb and 1.67–1.81 ppb, respectively)."

"During the CLP, the HONO concentration significantly decreased and remained at low level (0.28–0.66 ppb)."

"Due to nighttime formation and accumulation, the HONO concentration peaked (0.66 ppb) around midnight, then slowly decreased before sunrise."

"The HONO concentration did not show a significant increase during the morning rush hour (7:00–8:00 LT), indicating a substantial reduction in vehicle emissions during the CLP."

"The HONO concentration decreased to its minimum value (0.28 ppb) at 11:00 LT, then showed an increase around noon, reaching 0.33 ppb at 13:00 LT."

Line 233: add "the" before Beijing

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 251-253 in the revised manuscript:

"However, the CLP observed in this study, which occurred during the frequent winter haze pollution period (November 26 to December 23), differed significantly from the pollution situation in the Beijing urban area during the same period in other studies."

Line 261, 285, 287 and 292: I would add "period" after observation

Response: Thanks for your valuable comments. Revisions was made as the referee suggested. Lines 280-281, lines 306-307, lines 308-311 and lines 312-314 in the revised manuscript:

"To assess the impact of vehicle emissions in this observation period, the local emission factor EF_{emis} (=ΔHONO/ΔNOx) was derived based on ambient measurements."

"This indicated that the potential for heterogeneous conversion from NO_2 to HONO was stronger during this observation period, especially in DHP."

"Due to the absence of measurements of aerosol surface density (SA) in this observation period, $PM_{2.5}$ concentrations were used as a substitute to determine the impact of aerosols on the conversion of NO_2 to HONO at nighttime (Lu et al., 2018; Cai et al., 2017)."

"$HONO_{corr}$ exhibited a significant positive correlation with NO_2, with correlation coefficients (R^2) of 0.66, 0.45, and 0.38 during the DHP, PEP, and CLP, respectively, indicating that the heterogeneous reaction of NO_2 was an important source of HONO in this observation period."

Line 275: replace "cleaner period" with "cleaner periods"

Response: Thanks for your valuable comments. Revisions was made as the referee suggested. Lines 294-296 in the revised manuscript:

"Vehicle emissions accounted for 9.6 %, 11.7 %, and 17.6 % of nighttime HONO during the DHP, PEP, and CLP, respectively, indicating that the relative importance of direct emissions increases under cleaner periods, which was consistent with previous studies (Jia et al., 2020; Zhang et al., 2022c)."

Line 294: replace "was" with "were"

Response: Thanks to the referee for this comment. We carefully checked the sentence in Line 294: "...suggesting that the heterogeneous reaction of NO_2 on aerosol contributed to HONO formation during the DHP and PEP, which was closely associated with the higher $PM_{2.5}$ concentrations providing more reactive surfaces." Here, the antecedent of "which" is "the heterogeneous reaction of NO_2 on aerosol," which is singular. Therefore, the use of "was" is grammatically correct.

Line 296-297: remove the double references "Yan et al. (Yan et al., 2015) and Zhang et al. (Zhang et al., 2019b)"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Deleted the Yan et al. and Zhang et al. before the bracket. Lines 318-321 in the revised manuscript:

"For example, (Yan et al., 2015) and (Zhang et al., 2019b) reported that during haze pollution events in Beijing in the mid-2010s, the average $PM_{2.5}$ concentration could reach approximately 130 $\mu g\ m^{-3}$, with levels during severe haze episodes approaching 311 $\mu g\ m^{-3}$."

Line 320: replace "further" with "which"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 343-345 in the revised manuscript:

"The base model could only explain 4.2 %, 19.1 %, and 19.0 % of the observed HONO ($HONO_{obs}$) during the DHP, PEP, and CLP, respectively, which led to an underestimation of OH and O_3 concentrations in the atmosphere (Liu et al., 2019b; Tie et al., 2019)."

Line 379, 380, 412, 414, 418, 424, 428: I would add "the" before "three periods"

Response: Thanks for your valuable comments. Revisions was made as the referee suggested. Lines 407-408, lines 408-409, lines 442-443, lines 444-445, lines 448-450, lines 456-458, and lines 460-463 in the revised manuscript:

"Additionally, an extra HONO source, enhanced by photolysis and consuming OH radicals, was introduced for daytime HONO production during the three periods."

"The revised simulation results ($HONO_{sim,1}$) showed good agreement with $HONO_{obs}$, successfully reproducing the HONO variations during the three periods (Figure 5)."

"The HONO variation characteristics exhibited similarities and differences across the three periods."

"HONO exhibited similar diurnal variation trends in the three periods, with higher mixing ratios at night and lower during the day."

"The differences in pollutant concentration were related to the distinct HONO formation mechanisms and conversion frequencies during the three periods, reflecting the variations in atmospheric chemical processes."

"pNO_3 photolysis accounted for 12.7 %, 11.7 %, and 5.0 %, consistent with $PM_{2.5}$ concentrations in the three periods."

"Despite incorporating all known sources into the model, significant missing HONO sources remained during the three periods, accounting for 50.4 %, 16.9 %, and 7.0 %, respectively."

Line 397: Replace "declined" with "declines"

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 427-429 in the revised manuscript:

"The simulation results (orange scatter points in Figure 5) indicated that NOx reduction led to significant reductions in HONO levels, with declines of 42.7 % and 46.3 % during the DHP and PEP, respectively."

Line 419: Add "relative" before contribution since the absolute contribute is higher during the other two periods

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 451-453 in the revised manuscript:

"During the DHP and PEP, stronger correlations between nighttime HONO with $PM_{2.5}$ and NO_2 indicated a relative greater contribution from heterogeneous reactions, thereby reducing the relative impact of vehicle emission."

Line 421: Add “the” before dominant

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Lines 453-455 in the revised manuscript:

“The NO_2 heterogeneous reaction on ground was the dominant HONO source in all periods, contributing 45.5%, 37.8 %, and 44.0 % of simulated HONO, respectively.”

Line 431: “significantly reproduced” should be replaced by “significantly improved the agreement with”

Response: Thanks for your valuable comments. Revision was made as the referee suggested. Line 464 in the revised manuscript:

“Including these pathways in the model significantly improved the agreement with observed HONO.”

References

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