

Dear Reviewer,

We appreciate your careful consideration of our manuscript. We have carefully responded to all of your **point-by-point** comments and issues and have revised the manuscript accordingly. These revisions are described in detail below.

Reviewer #1

The authors have made commendable efforts to conduct a comprehensive field measurement in urban Beijing, spanning the unique transition period from the pre-pandemic COVID-19 era to the subsequent lockdown period. While the controversies outlined in the introduction section (lines 95-122) remain unresolved, this study observes a notable decrease in HONO concentration from the pre-pandemic eve to the lockdown phase. Furthermore, the study offers a reasonable explanation regarding the contributions of HONO sources. I find this work particularly valuable as it underscores the significance of vehicle emissions and highlights the rapid temporal shifts in the roles of HONO sources, which could be of great interest to readers of ACP. I recommend publication of the manuscript following minor revisions. Below are my specific comments.

Response: Thank you for your positive comments and good suggestions. We will respond to your comments point-by-point below.

1. The abstract requires improvement. Please remove lines 36-38 “which resulted in ... to measure the HONO and related pollutants”, and restructure the sentences for better clarity.

Response: Thank you for your suggestion. We have removed lines 36-38 “which resulted in the largest changes in air pollutant emissions in the history of modern atmospheric chemistry. A home-made Water-based Long-Path Absorption Photometer (LOPAP) along with other instruments were used to measure the HONO and related pollutants”. In the revised manuscript, we reorganized them into “**which resulted in a significant reduction in air pollutant emissions, providing a rare opportunity to understand the HONO budget in the atmosphere. We measured HONO and related**

pollutants” in lines 36-38.

2. Lines 67 and 80: The second and third paragraphs contain overlapping information. I recommend integrating the introduction about HONO concentrations into the first paragraph. Subsequently, the second paragraph should focus solely on direct emissions, while the third paragraph should discuss secondary formation.

Response: Thank you for your good suggestion. We have integrated the introduction of HONO concentrations into the first paragraph, the original “**Intensive studies have been carried out on HONO measurements and source analysis (Liu et al., 2020c; Liu et al., 2020d; Zheng et al., 2020; Zhang et al., 2020; Xue et al., 2020; Zhang et al., 2019a; Liu et al., 2019b). The concentrations of HONO in the atmosphere range from a few ppt in remote areas (Spataro et al., 2016) to several ppb, even several tens ppb in heavily polluted areas (Liu et al., 2019b; Liu et al., 2020c; Liu et al., 2020d; Zheng et al., 2020).**” in lines 63-67 has been moved to lines 61-66 in the revised manuscript.

As you suggested, the second paragraph only focuses on direct emissions in lines 68-79 in the revised manuscript “**Direct emissions include soils, biomass burning, vehicles, indoor air, and livestock farming. Soil emissions, which depend on soil types, microorganisms, water content, temperature, and pH (Kulmala and Petäjä, 2011; Weber et al., 2015; Kim and Or, 2019), are important sources of HONO. Biomass burning, often occurs in the summer and autumn when wheat/corn is harvested and wildfires are common (Zhang et al., 2019b; Sun et al., 2017; Sun et al., 2018; Peng et al., 2020). Vehicle emissions are considered an important source of HONO in traffic-intensive areas (Kramer et al., 2020; Li et al., 2021). This source is more important at nighttime compared with daytime (Zhang et al., 2016; Fu et al., 2019; Liu et al., 2020d). Recently, indoor emissions have also been proposed as a potential HONO source (Xue, 2022), which is related to the ventilation from high HONO concentrations in indoor air to low HONO concentrations in outdoor air (Zhang et al., 2019b). Livestock farming is a previously overlooked source of HONO, especially in agricultural areas.**”

The third paragraph discusses secondary formation in lines 80-88 in the revised

manuscript “Secondary formation of HONO includes gas-phase reaction between NO and OH radicals, photolysis of particulate nitrate, and heterogeneous reaction of NO₂ on ground and particulate matter surfaces, including photochemical heterogeneous reaction of NO₂. Gas phase reaction between NO and OH, photolysis of nitrate particles, and light-enhanced conversion of NO₂ are the main daytime sources of HONO (Liu et al., 2019c; Liu et al., 2020d; Zhang et al., 2022b). Furthermore, acid replacement processes may be a non-negligible source of daytime HONO in locations affected by soil-borne mineral dust deposition (Vandenboer et al., 2014). The heterogeneous reaction of NO₂ on various surfaces is widely regarded as an important source of HONO (Han et al., 2016; Liu et al., 2020b).”.

3. Line 71: Here, "harvest season" may not be the most accurate term. Biomass burning encompasses a range of activities, including wildfires, which can be particularly significant under certain circumstances.

Response: Thank you for your good suggestion. We have modified it in the revised manuscript to “Biomass burning, often occurs in the summer and autumn when wheat/corn is harvested and wildfires are common (Zhang et al., 2019b; Sun et al., 2017; Sun et al., 2018; Peng et al., 2020)” in lines 71-73.

4. Line 85: The authors omitted the acid displacement (VandenBoer et al. 2015, Liu et al. 2019)?

Response: Thank you for your good suggestion. In lines 85-86 in the revised manuscript, we added a sentence “Furthermore, acid replacement processes may be a non-negligible source of daytime HONO in locations affected by soil-borne mineral dust deposition (Vandenboer et al., 2014)”.

5. Line 79: Livestock HONO emission is negligible in urban Beijing. Similarly, the emission of HONO from soil, which may peak following fertilizer application, likely does not need to be considered.

Response: Thank you for your good suggestion. The HONO emissions from

Livestock in urban Beijing can be ignored, while soil emissions are slightly more important than Livestock. In this study, the contribution of the soil is even comparable to that of heterogeneous conversion of NO₂ on the surfaces of aerosol. Therefore, we still include the soil emissions when performing the budget analysis.

6. Line 263: heterogeneous yield of HONO instead of NO₂?

Response: Thank you for your good suggestion. In the revised manuscript in [line 262](#), we changed it to “**heterogeneous yield of HONO**”.

7. Section 3.1 Please present the result following the order of Figure 1 (a) ~ (i).

Response: Thank you for your good suggestion. In Figure 1, we moved the meteorological parameters to the end of this figure. And revised the description order of pollutants accordingly in [lines 317-340](#) in the revised manuscript.

8. Line 405: How about the heterogenous conversion on ground?

Response: Thank you for your good comments. The description here is inaccurate, in the revised manuscript, we corrected the sentence “These results imply that HONO might be more influenced by vehicle emissions than by heterogeneous reactions **on aerosol surfaces**” in [lines 404-406](#).

9. Line 235: Is HONO_{corr} equivalent to CHONO_{corr,t} as indicated in Eq. (6)? Please maintain consistency in the use of abbreviations.

Response: Thank you for your good comments. In Section 2.2, to maintain the consistency of the formula, we use “CHONO_{corr,t}”, and in subsequent chapters, for the convenience of readers, we use “HONO_{corr}”. In order not to mislead readers, in the revised manuscript, we use HONO_{corr} to represent it uniformly.

10. Line 507: In fact, Song et al. (2023) proposed two HONO production pathways distinct from the homogeneous reaction between NO and OH.

Response: Thank you for your good suggestion. The quote here is inappropriate, in lines 507-511 in the revised manuscript, we have modified it to “This is consistent with previous studies in urban Beijing (Gu et al., 2021; Jia et al., 2020; Liu et al., 2021). Interestingly, a recent study proposed a new mechanism through smog chamber experiments, that is, NO_x photooxidation (reaction of NO and adsorbed HNO₃) may be an important daytime HONO source (Song et al., 2023), although it has not yet been verified by field observations.”

11. Line 533: “a change of 25% and 95% of HONO sources, respectively.” What does this signify? It appears that the selection of the photolysis rate constant could have a significant influence.

Response: Thank you for your good comment. We agree with you that the selection of the photolysis rate constant has a significant influence on its contribution to the HONO source. The big difference in the estimated source here means both the upper and lower limits of the parameters might be improper. Thus, it requires a more restrictive method to evaluate the reasonability of the parameterization and assess the overall uncertainty. In our study, we further compared the estimated hourly HONO concentrations with the observed values to evaluate the parameters finally chosen. In lines 604-607 in the revised manuscript, we added a short paragraph “It should be noted that each source is sensitive to the corresponding parameter as discussed above. Thus, a more restrictive criterion is required to evaluate the reasonability of the parameterization. We further estimated the HONO concentration according to Eq. (2) and the parameters described in Sect. 2.2 to verify these calculated sources and sinks of HONO”. In addition, the overall uncertainty is 27.2% for the HONO budget evaluated based on Monte Carlo simulations. This has been pointed out in line 623 in the revised manuscript.

12. Lines 532-537: Move the results regarding soil emissions to the subsequent paragraph.

Response: Thank you for your good suggestion. We have moved the paragraph

“The yield of soil emissions in the P2 stage is also higher than that in the P1 stage due to the temperature rise in the P2 stage because the temperature will affect the soil emission flux (Oswald et al., 2013), while the importance of this source is negligible in this study. In M15 and M16, we amplify and shrink the soil emission flux by 10 times, respectively, and the change of the simulated HONO sources was less than 5%.” to the subsequent paragraph in [lines 553-558](#) in the revised manuscript.

13. Line 563: The NO₂ concentration decreased significantly from 26.9 ppb to 17.2 ppb (-36%) from P1 to P2, indicating a substantial reduction rather than a slight decrease.

Response: Thank you for your good suggestion. In [lines 562-564](#) in the revised manuscript, we revised it as “It is worth noting that the HONO formation rate from the heterogeneous conversion of NO₂ on the surface of aerosol does not decrease, which is caused by the increase in PM_{2.5} concentration along with a decrease in NO₂ concentration during the P2 period”.

14. Lines 569-572: It’s noteworthy that HONO exhibits minimal sensitivity to both the uptake coefficient (γ) and surface area concentration (A_s). However, the authors should provide an explanation for this phenomenon.

Response: Thank you for your good comments. According to eq(7), the contribution of the heterogeneous reaction of NO₂ on aerosol surfaces to the HONO source should be sensitive to both the uptake coefficient (γ) and surface area concentration (A_s). Although the absolute values change prominently as expected when the A_s or γ is increased or reduced, its relative contribution does not change obviously due to the small absolute value compared with other sources.

In [lines 574-578](#) in the revised manuscript, we have revised the paragraph “It should be noted that HONO is sensitive to the uptake coefficient and surface area concentration. When the uptake coefficient is expanded by 5 times or reduced by 10 times, the absolute HONO flux attributed to heterogeneous reactions increases 5 times or decreases 10 times, while the relative contribution is very low due to the small absolute value of heterogeneous reactions compared with other sources.”

15. Line 592: These variations (-9% to +40%) are significant and warrant attention.

Response: Thank you for your good comments. As mentioned in the previous reply, this sensitivity is closely related to the contribution of heterogeneous transformation of NO₂ at the ground surface to HONO. When the uptake coefficient is expanded by 5 times or reduced to 0.1, HONO changes by 40% and 9% respectively, indicating that HONO is sensitive to the NO₂ uptake coefficient on the ground surface, implying the importance of this source. In the revised manuscript, we added the sentence “**Indicating that HONO is sensitive to the NO₂ uptake coefficient on the ground surface.**” in lines 591-592.

16. Lines 621: The method utilized to estimate the overall uncertainty of the parameterization should be presented in the Experimental section.

Response: Thank you for your good suggestion. In lines 295-297 in the revised manuscript, we added a sentence “**Oracle Crystal Ball (version 11.1.2.4, Oracle's software for modeling, prediction, simulation, and optimization) (Rahmani et al., 2023) to evaluate the overall uncertainty of the parameterization through Monte Carlo simulations. The details are shown in Text S2 in the SI**”. In the revised SI, we added an introduction to the Monte Carlo algorithm as follows:

Text S2 Monte Carlo algorithm

The Monte Carlo algorithm is a method of estimating numerical values through random sampling. It can be used to estimate the overall uncertainty of the numerical value. A large number of samples are generated by random sampling from a probability distribution and the required numerical indicators are calculated based on these samples. Due to the limited number of samples, there is a certain error between the estimated value and the true value. We increase the number of sampling times to 10,000 to reduce statistical uncertainty.

When establishing the simulation model, the respective change ranges of the variables that affect HONO intensity are input, and the uncertainty of the modeling is evaluated by sampling from the probability distribution of the parameters to obtain the

overall uncertainty. In addition, the uncertainty of the model parameters is propagated to the model output through Monte Carlo sampling, and the uncertainty distribution of the results can be obtained. The formula for overall uncertainty can be expressed as:

$$\sigma = \sqrt{\frac{1}{N} \sum_{i=1}^N (x_i - \bar{x})^2}$$

σ represents the standard deviation of the overall uncertainty; N is the number of samples; x_i is the value of the i^{th} sample, and \bar{x} is the mean of the sample.

Reviewer #2

This paper presents an analysis of measurements of gas-phase nitrous acid (HONO), and related air pollutants/atmospheric chemical species, performed in the boundary layer in Beijing prior to and during the Covid-19 lockdowns in early 2020. The variation in abundance of HONO, its precursors and related species are used to quantify different chemical source and sink terms for HONO, and from their diurnal variation and change across the start of lockdown, inferences drawn re their relative importance – in particular direct vehicle emission.

The abundance and sources of HONO are very much a current topic in urban atmospheric chemistry/air pollution, as besides being a pollutant in its own right HONO is usually the major precursor to (reservoir for) the key oxidant OH. Lockdown presents a unique near-step-change opportunity to explore these processes. This paper adds to the developing understanding of these sources, and in that respect is a valuable contribution to the literature.

The measurements appear to have been carefully performed and are clearly presented, and the analysis is interesting, with clear signal in the change in mean diurnal variations (which are used to develop qualitative arguments).

However, I have substantial reservations about several key assumptions/aspects of the quantitative analysis approach, which in my opinion do not allow the authors to draw the conclusions they reach. These are:

Response: Thanks for your positive comments and good suggestions. We will reply to your concerns point-by-point below.

Major comments

1. Meteorology. Obviously, weather changes affect pollutant advection, import, dispersion, chemical processing, etc. The paper presents “before” vs “after” comparisons of concentrations either side of the start of lockdown, without consideration (beyond a basic vertical dispersion parameterization) of these effects. There is significant literature on the importance of correcting for meteorology—for example applying de-weathering approaches – several using the specific example of air

pollutant abundance in Beijing across lockdown (e.g. Jiabo et al., 2021; Shi et al., 2021; Lv et al., 2022). How much of the observed change (or lack of change) in each species is due to changes in the weather between the two time periods P1 and P2? It would be interesting to repeat the analysis using deweathered concentrations.

Response: Thank you for your good comments and suggestions. We agree with you that meteorology is always one of the important factors affecting the concentration of air pollutants. As you suggested, we performed the de-weather analysis using a Random Forest model with a deweather module.

In lines 344-368 in the revised manuscript, we added a paragraph “It is worth noting that changes in atmospheric pollutant concentrations are affected by both emissions and meteorology. Especially, during the lockdown period, meteorological conditions in Beijing were not conducive to the dispersion of pollutants, thus the impact of meteorological conditions on the concentration of these pollutants needs to be assessed. We use the random forest algorithm of machine learning to remove the influence of meteorology from air quality time series data by a deweather method. The details are present in Text S1 in the SI. The model performs well in predicting the concentrations of pollutants compared to the observations in both the training and test datasets (Table S5). The concentrations and relative changes of each pollutant after deweather are recorded in Table S6. After deweather, the mean concentration of PM_{2.5} increased significantly from 45.22±28.56 in P1 to 67.92±57.97 μg m⁻³ in P2 at a confidence level of 0.05, with an increase of 50.2%; The mean concentration of HONO was 0.89±0.37 ppb in P1, while it decreased to 0.51±0.25 ppb in P2, with a drop of 42.70%; The concentrations of NO and NO₂ significantly decreased from 15.44±18.40 and 23.28±7.28 ppb in P1 to 3.24±2.05 and 16.43±5.98 ppb in P2, respectively, which decreased by 79.02% and 29.42% respectively; SO₂ decreased from 2.27±0.69 in P1 to 1.48±1.18 ppb in P2, a decrease of approximately 34.8%; CO increased from 823.60±318.92 in P1 to 896±488.29 ppb in P2 (an increase of 8.79%) and O₃ increased from 16.98±5.62 to 22.60±4.10 ppb, an increase of about 33.1%, which was much lower than the change range of observed values (75.08%). As shown in Table S6, meteorological conditions have a significant impact on O₃ concentration. The impact

was +39.64% and +6.15% in P1 and P2, respectively. The impact of deweather on NO in the two periods was -16.18% and +32.79%, respectively. It was -13.75% and -4.81%, respectively, for NO₂. However, the changes of other species in the two periods after deweather fluctuated between 2.3% and 7.8%. This implies that meteorological conditions have an important impact on the concentrations of NO and O₃, while meteorological factors have little impact on HONO, SO₂, CO, and PM_{2.5}.”

In lines 420-422 in the revised manuscript, we added the sentence “After deweather, the HONO concentration decreased significantly from 0.89 ± 0.37 in P1 to 0.51 ± 0.25 ppb in P2 at a confidence level of 0.05, with a decrease of 42.7%. This means that meteorology has little impact on HONO.”

In the revised SI, we added the details about the random forest model to correct the influence of meteorology, including an introduction to the algorithm, a flow chart, and a model evaluation. The added content is as follows:

Text S1 De-weather model

Changes in atmospheric pollutant concentration are affected by emissions and meteorology. Machine learning models, including boosted regression trees and random forest (RF) algorithms, often exhibit higher predictive accuracy because of their advantages in modeling complex relationships between response variables and predictor variables (Zhan et al., 2018). By reducing the variance/bias and error of high-dimensional data sets, it has better performance compared to traditional statistical and air quality models. The algorithm resolves the relationship between air pollutant levels and their predictors, including meteorological parameters and time variables such as the day of the year (Julian Day), day of the week (Monday to Sunday), and hour of the day (0-23) (Grange et al., 2018). The input data set was randomly divided into a training data set for building the RF model (i.e., 70% of the input data set) and a testing data set (30% of the input data set) for testing the performance of the RF model using unseen data sets. The RF model is an ensemble model composed of many individual decision tree models (Breiman, 2001).

In the RF model, the bagging algorithm is utilized, which involves randomly selecting samples from the training dataset, with replacement, along with their

respective predictor features. Each decision tree is grown based on various decision rules that optimize the fitting between observed pollutant concentrations (response variable) and their predictor features. The selection of predictor features for each tree node is performed randomly to achieve the best possible split. The predicted pollutant concentrations are determined by aggregating the outcomes of all individual decision trees through a weighted average. The bagging process, by averaging predictions from bootstrap samples, helps reduce variance and mitigates overfitting issues in the model. As shown in Figure S1, the entire data set is randomly divided into two groups, one is the training data set, used to build the random forest model; the other is the test data set, used for testing without seeing the data set. The training data set accounts for 70% of the total data, and the rest is test data. Grange et al. (2018) built the RF model using the R “normalweather” package.

In our study, the parameters of the RF model are as follows: hourly concentrations of HONO, NO, NO₂, O₃, PM_{2.5}, SO₂, and CO as dependent variables, meteorological parameters (wind direction, wind speed, air temperature, humidity, and atmospheric pressure) and Time predictors (weekdays, hours) served as independent predictors. The training set uses randomly selected 70% of the data, and the remaining 30% is used as the test set. Random forest models were developed using the *rmweather* R package (Grange et al., 2018; Grange and Carslaw, 2019). The number of trees is 300, and the number of variables split in each node is 3. For each weather normalization, the explanatory variables are resampled (excluding the time variable) without replacement and randomly assigned to the dependent variable observations. The 1000 predicted values are then aggregated using the arithmetic mean to obtain the deweathered concentration.

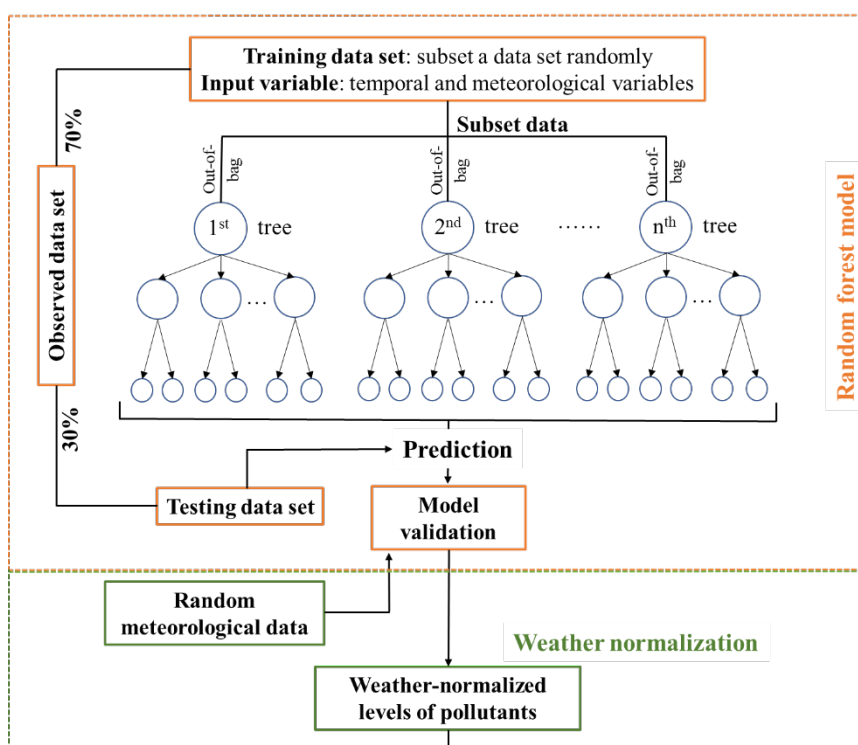


Fig. S1. The flowchart of the machine learning based RF algorithm.

Model performance evaluation

Evaluation metrics for the model can be found in Table S5. The random forest model showed good performance in predicting the data compared to the observations in the training and test datasets. Specifically, the R values range from 0.93-0.98. These extremely high correlation values indicate a strong relationship between the predicted values and the observed values, indicating that the characteristics of the established model are excellent. The FAC2 of each indicator is very small, indicating that our model meets the conditions for predicting scores. Likewise, lower NMB and NMGE values indicate that our model performs well. Through the verification of various indicators, it is believed that the model has good prediction ability.

Table S5. RF model performance for testing data set (in hourly time resolution).

Pollutants	RMSE	R	FAC2	MB	MGE	NMB	NMGE
HONO	0.21	0.93	0.86	0.01	0.15	0.02	0.21
NO	7.30	0.93	0.34	-0.21	3.76	-0.03	0.50
NO ₂	4.38	0.94	0.93	-0.04	3.12	0.00	0.16

O ₃	4.04	0.95	0.84	0.12	2.91	0.01	0.16
SO ₂	0.63	0.93	0.68	0.01	0.38	0.01	0.27
CO	164.55	0.96	1.00	4.22	114.60	0.00	0.13
PM _{2.5}	12.88	0.98	0.88	0.83	8.70	0.01	0.15

Note: FAC2 (fraction of predictions with a factor of two), MB (mean bias), MGE (mean gross error), NMB (normalized mean bias), NMGE (normalized mean gross error), COE (Coefficient of Efficiency), IOA (Index of Agreement).

In the revised SI, we have added Table S6(Table R2). After deweather, the concentration changes of different pollutants in the two time periods will be introduced in detail in the subsequent replies.

Table R2. Periods and concentration after deweather (mean \pm standard deviation) of PM_{2.5}, HONO, trace gases in field observation, and the percentages in parentheses are concentration changes after deweather. Relative change in observed values and deweather values in different periods.

Category	BCNY (1.1-1.24)		COVID (1.25-3.6)		Relative change	
	Deweather	Observed	Deweather	Observed	Deweather	Observed
PM_{2.5} ($\mu\text{g}/\text{m}^3$)	45.22 \pm 28.56 (-4.26%)	47.23 \pm 44.50	67.92 \pm 57.97 (-2.28%)	69.86 \pm 67.26	+50.20%	+47.91%
HONO (ppb)	0.89 \pm 0.37 (-8.25%)	0.97 \pm 0.74	0.51 \pm 0.25 (-3.77%)	0.53 \pm 0.45	-42.70%	-45.36%
NO (ppb)	15.44 \pm 18.40 (-16.18%)	18.42 \pm 29.24	3.24 \pm 2.05 (+32.79%)	2.44 \pm 5.40	-79.02%	-86.75%
NO₂ (ppb)	23.28 \pm 7.28 (-13.75%)	26.99 \pm 13.41	16.43 \pm 5.98 (-4.81%)	17.26 \pm 11.34	-29.42%	-36.05%
CO (ppb)	823.60 \pm 318.92 (-9.27%)	907.72 \pm 499.16	896 \pm 488.29 (-6.17%)	954.87 \pm 624.04	+8.79%	+5.19%
SO₂ (ppb)	2.27 \pm 0.69 (+8.61%)	2.09 \pm 1.36	1.48 \pm 1.18 (+0.68%)	1.47 \pm 1.95	-34.80%	+29.67%
O₃ (ppb)	16.98 \pm 5.62 (+39.64%)	12.16 \pm 10.79	22.60 \pm 4.10 (+6.15%)	21.29 \pm 11.78	+33.10%	+75.08%

2. Chemical lifetimes. The paper in effect performs a steady state analysis on HONO,

assuming any rate of change of concentration can be related to an imbalance in the in-situ source and sink terms. You can do this for short-lived species – such as OH – but only with care for species such as HONO, with lifetimes (these are midlatitude winter measurements) of tens of minutes. The measured HONO reflects the integrated chemical variability over the sampled air mass trajectory prior to its arrival at the measurement point. This will be heterogeneous – especially at ground level in the middle of a city (i.e. the OH and NO₂, etc will have varied a lot over the period of time – given by the HONO lifetime – that the air mass has traveled to the measurement point within the city). See arguments developed by Lee et al, JGR 2013, and related papers.

Response: Thank you for your good comments and suggestions. We agree with you that the measured HONO reflects the integrated chemical variability over the sampled air mass trajectory prior to its arrival at the measurement point. This might introduce uncertainty about the budget analysis using a steady state analysis. To confirm the possible affected areas, we carefully checked the representativeness of the dataset. Firstly, a potential source contribution function (PSCF) of HONO has been analyzed. Given a 10 min lifetime of HONO, the spatial distribution of HONO in the P2 is highly similar to that when compared with that in the P1 as shown in [Figure R2](#). This means air mass should have a similar impact on the HONO budget during these two periods. Secondly, we carried out deweather analysis using a random forest algorithm combined with a machine learning model. The details are shown in the SI. We found meteorology has little influence on HONO concentration during the whole observation period. This means HONO concentration is dominated by primary emissions and secondary formation. Thirdly, we compared the concentrations of HONO at BUCT and Institute of Atmospheric Physics (IAP) from Jan. 5th to Mar 24th when the data are available ([Figure R3](#)). The linear distance between these two sites is ~8 km. The HONO concentrations are comparable at these two sites. Thus, even though the concentration changes represent the integrated chemical variability over the sampled air mass trajectory prior to its arrival at the measurement point, it still represents the local feature in urban Beijing. Finally, the instrumentation time resolution of LOPAP was 6 s. We calculated the variation coefficient for the datasets with different time resolution, i.e., 1

h vs 6 s. A small variation coefficient of $\sim 0.02-0.05$ implies that a small uncertainty of HONO budget might be resulted from the lifetime of HONO. Thus, we think the possible uncertainty should not have a large influence on our conclusions when the budget is compared at a fixed site between two different periods.

In lines 205-214 in the revised manuscript, we added a short paragraph to clarify this possible uncertainty as “Given that the result of potential source contribution function (PSCF, Fig S2), the source distribution of HONO between BCNY and COVID was highly similar and the trend of HONO was similar (Pearson’s $r=0.78$) between BUCT and Institute of Atmospheric Physics (IAP, 8 km away from BUCT), the steady state analysis on HONO is applicable and reasonable even though the lifetime of HONO is several minutes in the atmosphere. In addition, the instrumentation time resolution of LOPAP was 6 s. We calculated the variation coefficient for the datasets with different time resolutions, i.e., 1 h vs 6 s. A small variation coefficient of $\sim 0.02-0.05$ implies that a small uncertainty of the HONO budget might result from the lifetime of HONO. Thus, we think the possible uncertainty should not have a large influence on our conclusions when the budget is compared at a fixed site between two different periods”. And we have added Figure R2 as Figure S2 in the revised SI.

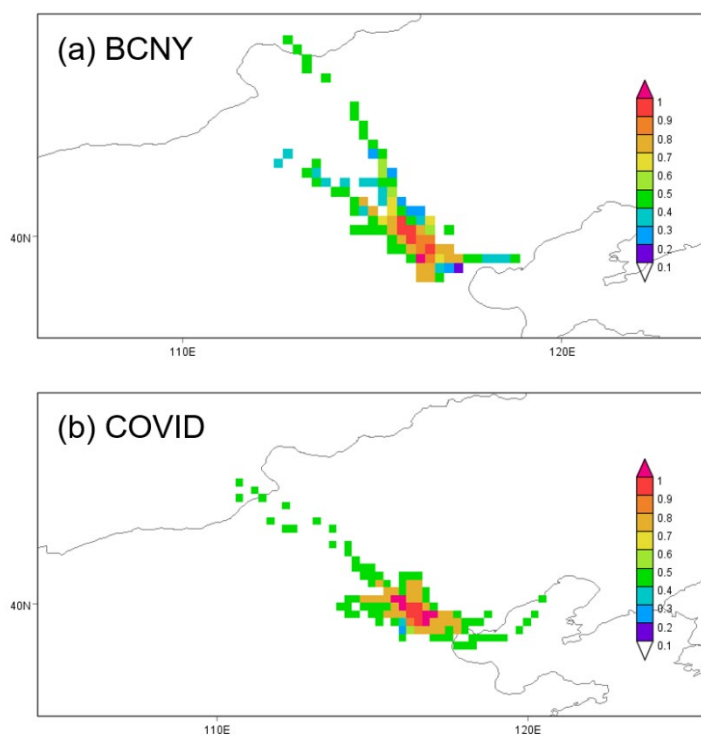


Figure R2. The potential source contribution function (PSCF) maps for the concentration of HONO (a and b are BCNY and COVID, respectively). The trajectory of the air mass is 12 hours.

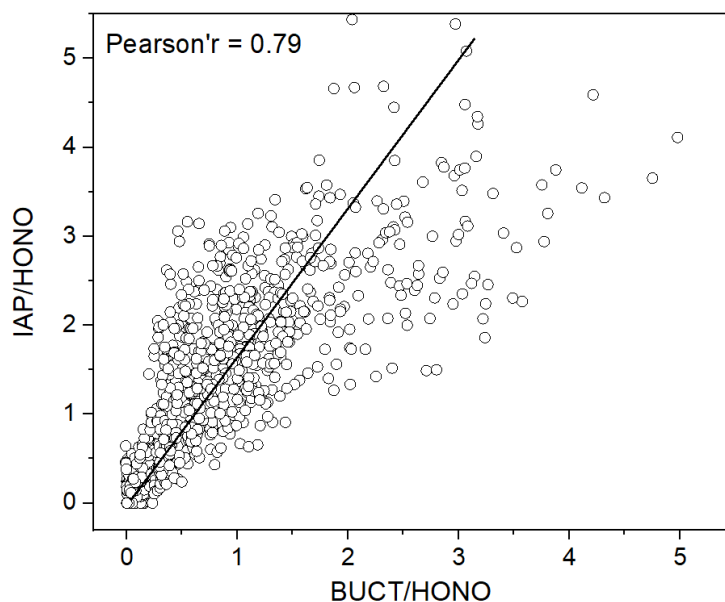


Figure R3. The comparison of HONO concentration between IAP and BUCT from Jan. 5th to Mar 24th.

3. Statistical significance/precision/uncertainty. The paper does not consider sufficiently the uncertainty in the (many) source and sink terms considered, and their propagation together (beyond the Monte Carlo result, which I can't believe includes the uncertainty in the individual inputs, e.g. OH concentrations). Is there any statistical power resulting from their combined uncertainties? Are the uncertainties given in the paper 1 or 2 standard deviations? Are differences statistically significant? The \pm ranges – even assuming these are 1 sd – suggest not (e.g. HONO/NO₂ ratio – changing from 0.038 ± 0.035 to 0.042 ± 0.034 – this is not a meaningful (statistically significant) change). There are several examples.

Response: Thank you for your comments and suggestions. As for your questions, we will reply one by one.

In our calculation, ten key parameters including source terms and sink terms were considered, and their respective fluctuation ranges were input, and then Monte Carlo was used to calculate the respective sensitivity distributions and uncertainties. In

addition, through comparison with field observations in the Beijing area, we found that our OH fitting performance is good. Of course, it should be noted that when the fluctuation range of various parameters were input, this may still bring some deviations although we have given full consideration and optimized based on existing research. Considering that the sources and sinks of HONO are complex and the selection of parameters in different studies may even differ by several orders of magnitude, our results should generally perform well.

For the comprehensive uncertainty, the uncertainty given in the paper is 1 standard deviation. In the revised manuscript, we added a description where it first appears “During P1, the measured concentration of PM_{2.5} varied between 0.2-288 $\mu\text{g m}^{-3}$ and the mean concentration was 47.2 ± 44.5 (mean $\pm 1\sigma$) $\mu\text{g m}^{-3}$.” in lines 317-318. And we added the sentence “The relative standard deviation is 27.2% for the HONO budget (details are in SI)” in line 623. In the revised SI, we added an introduction to the Monte Carlo algorithm. The added content is as follows:

Text S2 Monte Carlo algorithm

The Monte Carlo algorithm is a method of estimating numerical values through random sampling. It can be used to estimate the overall uncertainty of the numerical value. A large number of samples are generated by random sampling from a probability distribution and the required numerical indicators are calculated based on these samples. Due to the limited number of samples, there is a certain error between the estimated value and the true value. We increase the number of sampling times to 10,000 to reduce statistical uncertainty.

When establishing the simulation model, the respective change ranges of the variables that affect HONO intensity are input, and the uncertainty of the modeling is evaluated by sampling from the probability distribution of the parameters to obtain the overall uncertainty. In addition, the uncertainty of the model parameters is propagated to the model output through Monte Carlo sampling, and the uncertainty distribution of the results can be obtained. The formula for overall uncertainty can be expressed as:

$$\sigma = \sqrt{\frac{1}{N} \sum_{i=1}^N (x_i - \bar{x})^2}$$

σ represents the standard deviation of the overall uncertainty; N is the number of samples; x_i is the value of the i-th sample, and \bar{x} is the mean of the sample.

T-tests have been performed to confirm the significance of the differences mentioned in the manuscript and to the differences are statistically significant. For the \pm range that exists in the manuscript, such as “The HONO/NO₂ in the P2 period was higher than that in the P1 stage, especially in the daytime, although the values of HONO/NO₂ in both stages (P1: 0.036 \pm 0.016; P2: 0.041 \pm 0.038) were lower than that (0.052-0.080) reported by Cui et al (Cui et al., 2018). Subsequently, we further analyzed HONO_{corr}/NO₂ (details shown in Sect. 2.2). The HONO_{corr}/NO₂ attributed to secondary formation via heterogeneous reactions changed obviously after subtracting the interference of other HONO sources. As shown in Fig. S5, the daytime peak of HONO_{corr}/NO₂ in P2 became more prominent compared with that in Fig. 3e. At the same time, the HONO_{corr}/NO₂ (0.038 \pm 0.035) in P1 was slightly lower than that in P2 (0.042 \pm 0.034)” in the [lines 470-478](#) of the manuscript, it's still a statistically significant change at 0.05 level according to the T-test result.

Minor comments

I've noted some more points below but the authors need to address the points noted above vs the overall approach, to have confidence that the results of their analysis allow them to draw the conclusions presented in the paper.

1. Introduction – reviews different sources for HONO and their contribution, but this mixes together very different environments (i.e. the relative importance of different sources will vary for the measurement site vs a road tunnel vs a bare soil location vs the marine boundary layer vs livestock). Suggest distilling this to assess the key factors at the measurement location, ie city center.

Response: Thank you for your good suggestion. In [lines 67-137](#) in the revised manuscript, we revised the description as you suggested.

In the revised SI, we have added Table R2 as Table S1 and updated the sentence “The sources of atmospheric HONO consist of direct emissions and secondary

formation in the atmosphere. Direct emissions include soils, biomass burning, vehicles, indoor air, and livestock farming. Soil emissions, which depend on soil types, microorganisms, water content, temperature, and pH (Kulmala and Petäjä, 2011; Weber et al., 2015; Kim and Or, 2019), are important sources of HONO. Biomass burning, often occurs in the summer and autumn when wheat/corn is harvested and wildfires are common (Zhang et al., 2019b; Sun et al., 2017; Sun et al., 2018; Peng et al., 2020). Vehicle emissions are considered an important source of HONO in traffic-intensive areas (Kramer et al., 2020; Li et al., 2021a). This source is more important at nighttime compared with daytime (Zhang et al., 2016; Fu et al., 2019; Liu et al., 2020c). Recently, indoor emissions have also been proposed as a potential HONO source (Xue, 2022), which is related to the ventilation from high HONO concentrations in indoor air to low HONO concentrations in outdoor air (Zhang et al., 2019b). Livestock farming is a previously overlooked source of HONO, especially in agricultural areas.

Secondary formation of HONO includes gas-phase reaction between NO and OH radicals, photolysis of particulate nitrate, and heterogeneous reaction of NO₂ on ground and particulate matter surfaces, including photochemical heterogeneous reaction of NO₂. Gas phase reaction between NO and OH, photolysis of nitrate particles, and light-enhanced conversion of NO₂ are the main daytime sources of HONO (Liu et al., 2019b; Liu et al., 2020c; Zhang et al., 2022). Furthermore, acid replacement processes may be a non-negligible source of daytime HONO in locations affected by soil-borne mineral dust deposition (Vandenboer et al., 2014). The heterogeneous reaction of NO₂ on various surfaces is widely regarded as an important source of HONO (Han et al., 2016; Liu et al., 2020a).

Table S1 summarizes the sources of HONO at various locations. The type of observation site often has a great impact on the source intensity and contribution proportion of each source of HONO. In natural ecological areas or Antarctic stations with little human activity, the photolysis of nitrate is the main source of HONO during the day, and its contribution is much higher than the homogeneous reaction of NO and OH (Bond et al., 2023; Tang et al., 2024). In the ocean or areas close to the sea, the heterogeneous transformation of NO₂ becomes the main source of HONO, and the

transformation on the aerosol surface may be more important than that on the ground (Xing et al., 2023). In smoke collected near wildfires, it was found that the heterogeneous conversion contribution of NO₂ can reach 85%, making it the most important source of HONO (Chai et al., 2021). Emissions from soil and biological soil crusts are important in some areas where vegetation and soil are exposed (Meusel et al., 2018). For three different types of observation sites: rural, suburban, and urban, the relative importance of sources is also obviously different. In rural areas, there are usually no traffic activities, and are mainly affected by agricultural activities and animal husbandry, so traffic emissions can be ignored. During periods of intensive agricultural activity, soil emissions are the main source of HONO, accounting for up to 80% (Liu et al., 2019b). When there is little agricultural activity, the reaction of NO and OH and the heterogeneous transformation of NO₂ on the ground become the two main sources in rural areas (Xue et al., 2020; Song et al., 2022), accounting for up to 70%. In rural areas with developed animal husbandry, its direct emissions can contribute 39-45% of HONO (Zhang et al., 2023). Suburbs are mostly covered by vegetation, with a small number of villages nearby. The heterogeneous conversion of NO₂ is the main source of HONO, which can reach 70% of HONO sources (Fu et al., 2019; Ye et al., 2023). For highways, tunnels, and urban areas with heavy traffic, traffic emissions usually dominate HONO sources, accounting for 40% to 80% of HONO sources (Xu et al., 2015; Zhang et al., 2019c; Liu et al., 2020c; Kramer et al., 2020). In some ordinary urban areas where traffic activities are not so intensive, the heterogeneous conversion of NO₂ and the reaction of NO and OH are also the main sources of HONO in addition to traffic sources. It can be seen that the relative importance of different sources is often affected by the type of emission source near the observation site.

Although intensive studies have been performed on HONO sources, the contributions of different sources are still controversial (Zhou et al., 2011; Liu et al., 2014; Wu et al., 2019; Kramer et al., 2020; Meng et al., 2020). For the same type of observation area, the contribution of each source still diverges in different studies. For example, in mixed residential, commercial, and traffic areas, the importance of traffic emissions varies greatly. In some studies, it accounts for as much as 50% (Liu et al.,

2020c; Zhang et al., 2019a; Tong et al., 2016), while in some studies, it can be ignored (Zhang et al., 2020). A similar situation exists for the heterogeneous conversion of NO₂. Some studies suggest that this process is not important (Tong et al., 2015; Zhang et al., 2019c; Zhang et al., 2022), while some studies believe that it can contribute at least 70% of HONO (Meng et al., 2020; Zhang et al., 2020; Jia et al., 2020). It should be noted that the contribution of NO₂ heterogeneous reaction to HONO greatly depends on the choice of NO₂ uptake coefficient (γ_{NO_2}), which varies from 10⁻⁸ to 10⁻⁴ in different studies (Meng et al., 2020; Liu et al., 2020a; Ge et al., 2019; Liu et al., 2015; Liu et al., 2020c). Vehicle emissions also have similar characteristics because the HONO emission rate strongly depends on the emission factor, i.e. the ratio of HONO/NO_x (Kramer et al., 2020; R. Kurtenbach et al., 2001; Zhang et al., 2019c), which ranges from 0.03% to 2.1% (Liao et al., 2021). For other HONO sources, the relative importance is affected by many parameters, such as reaction kinetics for photolysis of nitrate, OH concentrations for homogeneous reaction between NO and OH, emission fluxes for soil emissions, and so on. Thus, the HONO budget still has a large uncertainty. In particular, it is an open question how to prove the importance of a specific reaction pathway or a source of atmospheric HONO.”

Table R2. Summary of HONO observation sites and source contributions

Location (References)	Date	Measurement area	Site situation	Source contribution
Antarctica (Bond et al., 2023)	2022.01	Research Station	Clean area, covered with ice and snow	Photolysis of nitrate in snow is very important, and its contribution is 10 times greater than the reaction between NO and OH.
Shenzhen (Tang et al., 2024)	2019.10	natural ecological area	Along the coast, there are fewer human activities and more vegetation.	Photolysis of large amounts of nitrate in coarse particles completely compensates for unknown sources during the daytime (66%).
China (Xing et al., 2023)	2018.05	sea edge	coastal	In inland areas, the NO ₂ heterogeneous reaction on the ground is more important; in coastal and ocean cases, the contribution of aerosol surfaces is greater.
Idaho (Chai et al., 2021)	2018.08	wildfire zone	Smoke collected near five wildfires	In the aging smoke during the daytime, the heterogeneous conversion of NO ₂ reaches 85%, followed by NO+OH.
Guangzhou (Li et al., 2012)	2006.07	rural area	Close to farmland, low traffic emissions	The main source at night is NO+OH and the heterogeneous conversion of NO ₂ on the ground, and traffic can be ignored
Cyprus (Meusel et al., 2018)	2016.04	rural area	Along the coast, a lot of vegetation is exposed	Emissions from soil and biological soil crusts are important.
Melpitz (Ren et al., 2020)	2018.04	rural area	Nearby are meadows, agricultural areas, and forests	Nocturnal HONO: Heterogeneous conversion of ground NO ₂ dominates, and traffic emission is a secondary source.
Wangdu (Liu et al., 2019b)	2014.06	rural area	Intensive agricultural activities and no traffic emissions	Noonday HONO: Soil emissions account for 80%.
Wangdu	2017.12	rural area	No traffic emissions,	Noonday HONO: The heterogeneous conversion of NO ₂ on

(Xue et al., 2020)			surrounded by farmland	the ground is 36%, NO+OH is 34%, and the others can be ignored.
Wangdu (Song et al., 2022)	2020.06- 2020.09	rural area	No traffic emissions, surrounded by farmland	Noonday HONO: The heterogeneous conversion of NO ₂ on the ground is dominant (43-62%), followed by NO+OH 12-38%, and the rest are less than 5%.
Wangdu (Zhang et al., 2023)	2020.09- 2021.08	rural area	Seriously affected by agriculture and animal husbandry	Direct emissions from rural areas, including animal husbandry, account for 39-45% and cannot be ignored.
Hongkong (Zhang et al., 2016)	2011.08	suburbs	Near the airport, surrounded by vegetation and close to the South China Sea	The heterogeneous conversion of NO ₂ on the ground is 42%, soil emission is 29%, marine source is 9%, NO+OH is 6%, aerosol surface conversion is 3%, and traffic is 2%.
Hongkong (Xu et al., 2015)	2011.08- 2012.05	suburbs	Areas near airports and highways are mostly covered by vegetation.	Nocturnal HONO: Traffic dominates in the first half of the night (59%), and the heterogeneous conversion of NO ₂ on the ground dominates in the second half of the night.
Heshan (Fu et al., 2019)	2017.01	suburbs	Lots of vegetation and farmland, with some scattered villages	Heterogeneous conversion of NO ₂ is 72%, traffic is 8%, and NO+OH is 3%. Noonday HONO: Photolysis of nitrate accounts for more than 50%.
Taizhou (Ye et al., 2023)	2018.06	suburbs	Borders farmland and fish ponds	Noonday HONO: The heterogeneous conversion of NO ₂ on the ground is 71%, followed by NO+OH, traffic, and aerosol surface conversion. Nocturnal HONO: Heterogeneous conversion of NO ₂ on the ground is dominant (55%).
Beijing (Tong et al., 2015)	2014.11	urban area suburbs	Densely populated and busy with traffic By the lake, with farmland	Nocturnal HONO: Traffic emission is 40%, NO+OH is 42%, and others are 18%. Nocturnal HONO: Traffic emission is 8%, NO+OH is 11%,

Location	Year	Area	Environment	Findings
Beijing (Tong et al., 2016)	2014.12	urban area suburbs	nearby	and others are 81%。
Beijing (Zhang et al., 2019a)	2006.08	urban area	Densely populated and busy with traffic By the lake, with farmland nearby	Nocturnal HONO: Traffic emission is dominant (49%), and the reaction of NO and OH is also important. Nocturnal HONO: Heterogeneous conversion of NO ₂ is the main source, and traffic is 10%.
Beijing (Zhang et al., 2019c)	2016.12	urban area	Mixed residential, commercial, and transportation area	Nocturnal HONO: Traffic is 41%, ground heterogeneous conversion is 27%, and aerosol surface conversion is 20%. Nocturnal HONO: Traffic emission is dominant, reaching 52%, and heterogeneous conversion is not an important pathway.
Beijing (Meng et al., 2020)	2016.12	urban area	Densely populated and busy with traffic	High altitude during haze: HONO is dominated by heterogeneous conversion on the aerosol surface; Near the ground: Heterogeneous conversion of NO ₂ on the ground is dominant, followed by traffic, accounting for 29% Nocturnal HONO: The light-induced heterogeneous transformation of NO ₂ on the ground is dominant, and aerosol surface conversion can be ignored.
Beijing (Gu et al., 2021)	2017.05 2018.01	urban area	Mixed residential, commercial, and transportation area	Nocturnal HONO: Traffic emission is dominant, reaching 50%, and heterogeneous conversion is not an important pathway. Nocturnal HONO: NO+OH is dominant.
Beijing (Liu et al., 2020c)	2018.02- 2018.07	urban area	Mixed residential, commercial, and transportation area	Nocturnal HONO: Nitrate photolysis and NO+OH are important.

Beijing (Zhang et al., 2020)	2018.04	urban area	Mixed residential, commercial, and transportation area, 325m vertical observation.	At different altitudes, the heterogeneous conversion of NO ₂ is the most important source, accounting for more than 70%. Among them, the aerosol surface is dominant.
Beijing (Jia et al., 2020)	2018.08	urban area	Mixed residential, commercial, and transportation area	Traffic is 18%, NO+OH is 31% (clean) and 7% (haze), and the aerosol surface conversion can reach up to 88%, which is very low on the ground. Nitrate photolysis is 15%, Noonday HONO: NO+OH is 22%, traffic is 19%, and
Beijing (Liu et al., 2021)	2018.06	urban area	Mixed residential, commercial, and transportation area	Heterogeneous conversion on the aerosol surface is 19%.
	2018.12			Noonday HONO: Heterogeneous conversion on the aerosol surface is 30%, Heterogeneous conversion on the ground is 25%, and traffic is 20%.
Beijing (Zhang et al., 2022)	2019.01	urban area	Densely populated and busy with traffic	Traffic is 28%, Heterogeneous conversion on the ground is 27%, and aerosol surface conversion is 15%.
Beijing (Li et al., 2021b)	2019.06	urban area	Mixed residential, commercial, and transportation area	Nocturnal HONO: The heterogeneous conversion of NO ₂ is the main pathway, followed by NO+OH. Traffic is 30%.
Shijiazhuang (Liu et al., 2020b)	2019.12- 2020.03	urban area	mixed traffic and residential area	Nocturnal HONO: The heterogeneous conversion of NO ₂ on the ground is dominant, followed by aerosol surface conversion.
Beijing-Tianjin- Hebei (Zhang et al., 2019b)	2017.12	urban area	Less traffic emissions and intensive agricultural activities	Nocturnal HONO: Traffic and heterogeneous conversion of NO ₂ are the main sources.
Xi'an	2015.08	urban area	Mixed residential, commercial,	Nocturnal HONO: The heterogeneous conversion of NO ₂ is

(Huang et al., 2017) Shanghai (Cui et al., 2018)	2016.05	urban area	and transportation area Mixed residential, commercial, and transportation area	the main pathway, followed by NO+OH. Traffic is 19%. Nocturnal HONO: Heterogeneous conversion of NO ₂ is the main source.
Nanjing (Zheng et al., 2020)	2015.12	urban area	To the west of the steel plant and petrochemical refinery, 15 kilometers from the city center	The heterogeneous conversion of NO ₂ is dominant, accounting for 50%, and traffic is 11%.
Nanjing (Liu et al., 2019a)	2017.11- 2018.11	urban area	Mixed residential, commercial, and transportation area	Traffic is 23%, heterogeneous conversion on the ground is 36%, Soil emissions can reach 40% in July and August. The aerosol surface conversion reaches 40% (severe haze periods).
Changzhou (Shi et al., 2020)	2017.04	urban area	Mainly residential and commercial areas, with no roads and industrial activities,	Nocturnal HONO: Heterogeneous conversion of NO ₂ is 54%, traffic is 32%, and NO+OH is 14%. Noonday HONO: Nitrate photolysis is important.
Guangzhou (Yu et al., 2022)	2018.10	urban area	mixed traffic and residential area	Nocturnal HONO: The three main sources are the heterogeneous conversion of NO ₂ on the ground, traffic, and NO+OH. The aerosol surface conversion and soil emissions are not important.
Birmingham (Kramer et al., 2020)	2016.11	urban area	road tunnel	Traffic is dominant, accounting for 66% (up to 86%), the heterogeneous conversion of NO ₂ is only 5%,

2. L174 – NO₂ measured by 42i – selectivity for NO₂, the NO₂ data will include other NO_y species (including HONO).

Response: Thank you for your good comments. This is taken into account in our observations, so in the calculations, the NO₂ concentration has been subtracted from the HONO concentration. In lines 173-174 in the revised manuscript, we added a sentence “Notably, the NO₂ measured by 42i includes HONO. Thus, it has been corrected”.

3. L256 how was $j_{(\text{HONO})}$ calculated

Response: Thank you for your comments. $j_{(\text{HONO})}$ is simulated in MCM using $j_{(\text{NO}_2)}$ data observed at our site. The details are as follows: In each case, variation of photolysis rate with solar zenith angle can be described well by an expression of the following form,

$$J = l (\cos \chi)^m \exp(-n \cdot \sec \chi)$$

by optimizing the values of the three parameters, l , m , and n are the parameters of the reaction respectively, χ is the solar zenith angle (Saunders et al., 2003). In line 256 in the revised manuscript, we added a sentence “ J_{HONO} is simulated in a box model using J_{NO_2} data observed at our site”.

4. L189 heterogeneous reactions of NO₂ have been accounted for – I didn’t follow this section, may need rewording.

Response: Thank you for your suggestion. I am sorry for confusing you. In the revised manuscript, we made changes and deleted the “have been accounted for” in “The sources including vehicle emissions (E_{vehicle}), soil emissions (E_{soil}), the reaction of NO and OH ($P_{\text{NO-OH}}$), the photolysis of particulate nitrate (P_{nitrate}), and the heterogeneous reaction of NO₂ (P_{aerosol} and P_{ground})” in lines 189-191.

5. L217 what is the conversion factor alpha?

Response: Thank you for your good suggestion. It is a unit conversion factor of emission flux from $\text{g m}^{-3} \text{ s}^{-1}$ to ppb h^{-1} . In lines 217-218 in the revised manuscript, we update the “conversion factor (α , from $\text{g m}^{-3} \text{ s}^{-1}$ to ppb h^{-1})”.

6. L227 using NO₂ and CO – NO₂ concentrations will vary with a lifetime of a minute

or so (with respect to the NO_x-O₃ PSS) and 6-12 hours or so (with respect to NO_x removal). CO concentrations will vary with a lifetime of several weeks. Is it valid to use both in the same in situ emission analysis?

Response: Thank you for your good suggestion. When calculating k_{het} , equation 3 does not consider the impact of the boundary layer on the concentration of pollutants. CO is considered to be a relatively stable substance and is usually used for partially eliminating the variation of the boundary layer height. Thus, equation 4 corrects the boundary layer variation by normalizing to CO concentration. This method has been widely used to calculate the heterogeneous uptake kinetic of NO₂ on aerosol in previous studies (Zhang et al., 2020; Zhang et al., 2019c; Li et al., 2012). It is expected to get a more accurate k_{het} because the influence of the variation of boundary layer height has been accounted for. It is worth noting that during a year of HONO observations in coastal cities in southeastern China (Hu et al., 2022), the author compared the results of balancing NO₂, CO, and BC and found that CO performed well, and finally selected CO as the parameter for k_{het} calculation.

In lines 232-234 in the revised manuscript, we have pointed out that “To decrease the contribution of boundary layer height variation on the k_{het} calculations, we normalized HONO concentration to CO concentration as the same as reported in the literature (Zhang et al., 2019c; Li et al., 2012)”.

7. L256 / table 1 – what is the estimated OH concentration & how does it compare with measurements – the values in L436 (presumably 24 hour mean, around 4-7e5) seem much lower than those observed in wintertime Beijing (2.7e6 as 24-hour mean; Slater et al., 2020).

Response: Thank you for your good suggestion. The article by Slater et al. in 2020 describes it as follows: “Averaged over the full observation period, the mean daytime peak in radicals was 2.7×10^6 , 0.39×10^8 , and $0.88 \times 10^8 \text{ cm}^{-3}$ for OH, HO₂, and total RO₂, respectively”. Our estimated 24-hour mean concentration of OH ranges from 1.1×10^5 to 2.5×10^6 , which is very close to the daytime peak observed in Beijing in winter (2.7×10^6) (Slater et al., 2020). The daily mean value ($4-7 \times 10^5$) is therefore lower than the maximum value at noon. As shown in Figure R4 or S3, the estimated OH concentrations are in good agreement with the observations in Beijing.

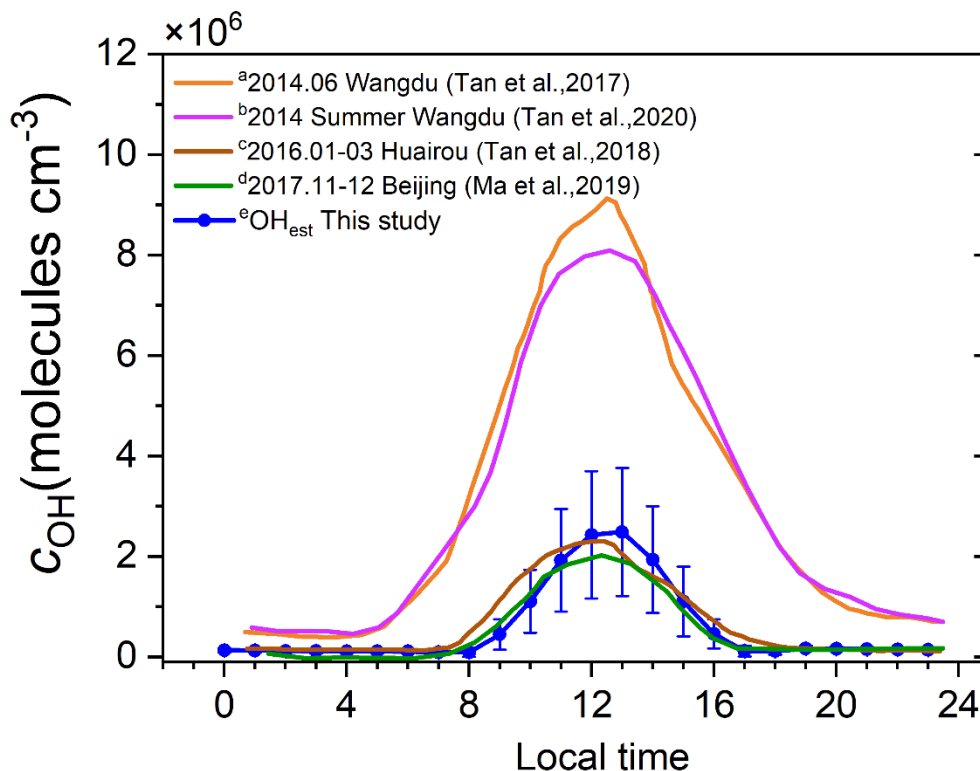


Figure R4. Diurnal variation of OH concentrations observed in different areas of the North China Plain (a-d) (Tan et al., 2017; Tan et al., 2018; Ma et al., 2019; Tan et al., 2020) and parameterized fitting in this study (e).

8. L269 Parameterisation of OH concentrations. We might expect that the main source of OH in Beijing is $\text{HO}_2 + \text{NO}$, the main sink for OH is $\text{OH} + \text{NO}$, and the main primary source of OH is HONO photolysis (one can then argue about if HONO is acting as a primary source or a reservoir). The parameterization given was developed for rural sites (as the authors note), where NO levels would be much lower, and was developed prior to more recent understanding of HONO abundance (it is over 20 years old). Is it valid to use? L286 – considering these above I do not agree that we can be optimistic about the estimated OH concentrations.

Response: Thank you for your good suggestion. Notably, this parameterization scheme was developed based on measurements at rural sites (Ehhalt and Rohrer, 2000), where NO_x concentrations were lower than in urban environments. Alicke et al. (Alicke, 2002) found OH concentrations estimated with this scheme were in good agreement with those calculated according to a pseudo-steady state method during the pollution period in urban environments (such as Milan) although some uncertainty was expected.

In our previous study (Liu et al., 2020d), we also found that the estimated OH concentrations using this method were comparable with those observed values in the North China Plain (Tan et al., 2019). Thus, daytime OH concentrations estimated using this method should be overall credible although the uncertainty is inevitable. Fig R2 (Fig S2) summarizes the observed OH concentrations in the North China Plain. The results estimated in this study are slightly lower than those observed in Wangdu, but almost consistent with those in Beijing and Huairou. In summary, although the parameterization was developed many years ago, judging from the performance results, it still has excellent performance in fitting OH.

In lines 283-286 in the revised manuscript, we have highlighted the points “The results estimated in this study are slightly lower than those observed in Wangdu (Rural), but almost consistent with those in Beijing (Urban) and Huairou (Suburb). In summary, we should be optimistic about the estimation of OH concentration.”

9. L369 – PM_{2.5} components increased obviously in P2 vs P1 – from the plot it is not obvious to me that they do: is there a statistically significant change? What about changes in the meteorology?

Response: Thank you for your comment. The increasing trend in Figure 1 is not very obvious, and we further summarize their concentrations in P1 and P2 in Table S4. In the revised manuscript, we added the sentence “It can be seen from Figure 1 and Table S4 in SI. All the major components of PM_{2.5}, including sulfate, nitrate, ammonium, chloride, and organic aerosol, increased obviously in P2 compared to P1.” in lines 369-371. The difference in PM_{2.5} between the two periods is statistically significant ($P < 0.05$). In the revised manuscript, we added the sentence “The PM_{2.5} concentration after deweather increased significantly from 45.22 ± 28.56 in P1 to $67.92 \pm 57.97 \mu\text{g m}^{-3}$ in P2 at a confidence level of 0.05, with an increase of 50.2%.” in lines 352-354.

10. L409 the traffic index data are useful. Consider showing P1/P2 on these plots. Consider changing the box/whisker plot to linear (not log) – this will assist the reader to follow which differences are statistically significant.

Response: Thank you for your good comment. We have changed the logarithmic plot of Figure 2b into a linear plot.

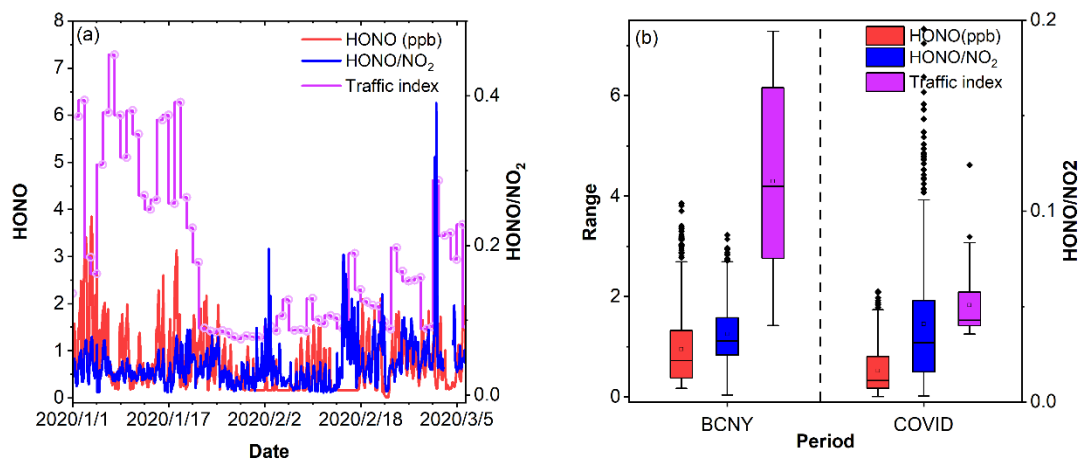


Figure R2. (a) Times series of HONO, traffic index, and HONO/NO₂, (b) Box plots of HONO, HONO/NO₂, and the traffic index in Beijing during different periods (BCNY=P1, LOCK=P2).

11. concentrations of NO changed: You cannot conclude this without a statistical test, esp for NO₂ and HONO.

Response: Thanks for your suggestion. Regarding the decrease in NO concentration, we did a T-test and found that there was a significant difference in the concentrations between the two time periods ($P < 0.05$). In lines 587-588, it has been pointed out as “This can be explained by the significant decrease ($P < 0.05$) in NO and direct emissions of HONO from traffic.”

12. L448 Figure 3 the shift in diurnal profiles is interesting, explore further (esp panel 5, HONO/NO₂)?

Response: Thanks for your suggestion. The daily variation curves can provide us with a lot of information. For example, HONO, NO_x, SO₂, and O₃ show similar diurnal variations, while the concentrations of HONO, NO_x, and SO₂ decrease obviously during the COVID lockdown when compared to that before the COVID lockdown and O₃ concentrations increase. However, the diurnal curves of HONO/NO₂ and PM_{2.5}×NO₂ show obvious daytime peaks during the COVID lockdown, which are different from those before the lockdown. In addition, their nocturnal values are comparable between the two periods, while their daytime values are higher during the COVID lockdown than those before the lockdown. Because both HONO/NO₂ and PM_{2.5}×NO₂ are indicators for the heterogeneous reaction of NO₂ on aerosols to form HONO, these results imply that the heterogeneous reaction rate during the COVID lockdown should

be higher than that before the lockdown. However, the absolute concentrations of HONO during the COVID lockdown are lower than those before the lockdown. It suggests that heterogeneous reactions should be an unimportant source of HONO in Beijing.

In lines 484-486 in the revised manuscript, we emphasize this point “**In summary, we propose that during our observation period, heterogeneous reactions of NO₂ should have a relatively minor contribution to HONO production**”.

13. L478 contribution (not interference) of other HONO sources. This matters vs literature discussion of interferences in some HONO measurement approaches.

Response: Thanks for your good suggestion. In our HONO_{corr} calculation method, primary and other secondary sources of HONO not attributing to heterogeneous reactions have been subtracted. Thus, the obtained value is more reasonable to present a heterogeneous conversion of NO₂.

To make it clearer, we revised the sentence in lines 477-478 in the revised manuscript as “The HONO_{corr}/NO₂ attributed to secondary formation via heterogeneous reactions changed obviously after **subtracting other secondary HONO sources**”.

14. L402 ifs there any info on changes in fleet composition? Big change in total traffic, but the greatest change may be in discretionary journeys (private cars) while deliveries etc(potentially much greater per-vehicle emitters) may have continued.

Response: Thanks for your suggestion. Unfortunately, changes in fleet composition are unavailable. The situation you mentioned may exist, that is, some delivery trucks still have continued. It should be noted that if the HONO emissions by trucks along with the heterogeneous reaction sources increase during the lockdown, an increase in HONO concentrations should be observed. Thus, we ascribe the reduction of HONO concentrations to the decrease in total emissions of HONO from vehicles as supported by the decreases in the traffic index. Therefore, even if the emissions of trucks increase due to the load, this increase is still insignificant compared to the overall emissions of vehicles before the lockdown.

15. L611 the agreement is good to see but is there really confidence in the combined uncertainty of the terms entering the calculation – especially [OH] – to have confidence? L624 same point.

Response: Thanks for your good suggestion. Your opinion is very reasonable. Although we have calibrated each parameter strictly and as accurately as possible, we still cannot guarantee 100% accuracy. For example, for the fitting of OH radicals, although our fitting agrees well with the observations in Beijing, this still does not mean that our results are 100% correct. Considering the complex sources of HONO, uncertainty in the calculation is inevitable, while it is acceptable. In particular, the estimated hourly HONO concentrations are well in agreement with the observed ones. There are 27.2% in total. This means that the simultaneous equations (6 sources and 4 sinks) have been verified 10,000 times. To our best acknowledgment, this is the first time to constrain the parameterization for HONO source budget analysis based on steady-state analysis.

16. L637 plus – these changes must be considered in the context of (potential) changes in meteorology between the two periods – it can be very misleading to simply compare means calculated from two different, fairly short, date periods.

Response: Thanks for your good suggestion. Your suggestion is great. Changes in atmospheric pollutant concentration are affected by emissions and meteorology. The concentrations and relative changes of each pollutant after deweather are recorded in Table R2 (Table S6).

In the revised manuscript, we added the sentence “**After removing meteorological factors, the change proportions of PM_{2.5} concentration in the two stages were -4.26% and -2.28% respectively. The HONO changes were -8.25% and -3.77% respectively, the CO changes were -9.27% and -6.17% respectively, and the SO₂ changes were +8.61% and +0.68% respectively. The change proportions are all less than 10%, which means that the impact of changes in meteorological factors on PM_{2.5}, HONO, CO, and SO₂ is very weak. However, the change proportions of NO in the two stages were -16.18% and +32.79%, respectively, and O₃ was +39.64% and +6.15% respectively. The change ratio is greater than 30%, indicating that NO and O₃ are greatly affected by meteorology. In addition, the changes in NO₂ were -13.75% and -4.81% respectively, implying that NO₂ is also affected by meteorological factors. From the entire observation period, except for O₃, the changes of other species in the two periods fluctuated between 2.3% and 7.8% after deweather, all less than 8%. In general, after removing the meteorological effects, NO increased by 79%, NO₂ increased by approximately 29%, HONO decreased by approximately 43%, and PM_{2.5} increased by approximately 50%. It is worth noting**”

that O₃ increased by about 33%, which is much lower than the change in observed values (75.08%) (as shown in Table S6).” lines 638-652.

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