

Dear Editor,

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

Editor's comments

The revised manuscript was reviewed again by Referee #2, who is now largely satisfied with the answers and revisions. Before the manuscript can be published, some more clarifications are needed as suggested by the reviewer.

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

1. In particular, the HONO source contributions in the abstract should be provided with error bars (e.g., nighttime vehicle emissions of 53% +/- ?), and in the conclusions, it should be mentioned which of the assumed parameters in the budget analysis cause the largest uncertainties in the estimated HONO source and how much.

Response: Thank you for your good suggestions. In the revised manuscript, we have added error bars for contributions from different sources of HONO. Through sensitivity analysis, we found that the source of HONO is most sensitive to OH radicals and $J_{\text{NO}_3^-}$. The uncertainty caused by OH radicals is $\pm 5\%$ using the observed values reported in the literature as constraints. For $J_{\text{NO}_3^-}$, there is a lack of direct observational data. Although parameter optimization was carried out in this study in combination with the results of the literature, it still brought the greatest uncertainty to the HONO source and sink assessment, with a value of $\pm 19\%$.

In lines 694-695 in the revised manuscript, we added a sentence “**Through uncertainty assessment, it was found that the assumption of $J_{\text{NO}_3^-}$ would have the greatest uncertainty, with a standard deviation of $\pm 19\%$. Nevertheless, this** study confirms that reducing anthropogenic emissions can indeed reduce the concentration of HONO in the atmosphere.”

2. In Table S3, a definition of the 'Sensitivity' (last column) is missing.

Response: Thank you for your good suggestions. In Table S3 in the revised SI, we

added the sentence “The source of HONO is affected by many factors, and its concentration varies with any one of these factors. The sensitivity here is calculated by univariate analysis, that is, observing the changes in HONO concentration by changing only one variable but with all other variables unchanged,”

3. Some numbers in the conclusions sections are provided with unreasonable precisions (e.g. " HONO changes were -8.25% and -3.77%"). Please correct.

Response: Thank you for your good suggestions. In the revised manuscript, we have modified the significant figures of the data. For example, “HONO changes were -8.25% and -3.77%” was changed to “HONO changes were -8.3% and -3.8%”.

Reviewer #2

I am grateful to the authors for the extensive and detailed work to address the comments raised previously – these substantially improve the manuscript. In particular, the application of deweathering approaches to separate the meteorological changes from emission changes in assessing the pre/post lockdown concentrations significantly increases the robustness of the inference regarding changes in HONO due to emissions.

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

1. I am still slightly unclear if the (change in) deweathered concentrations are used in the subsequent analysis, or the raw concentrations – with implications for the emissions sources following on – maybe needs to be clarified explicitly.

Response: Thank you for your good suggestions. We conducted the budget analysis of HONO using the deweathered concentrations as shown in Figure R1 (or Figure S10). The corresponding results are shown in Figure S9 (Figure R2) using the raw observed concentrations. The daytime source contributions of HONO almost did not change in both cases (using raw data or deweathered data) whatever the periods. However, the nocturnal contribution of vehicle emissions increased from 53% with the raw dataset to 63% with the deweathered dataset, along with a decrease of heterogenous reactions on ground surface from 31% to 19% before the Chinese New Year. It also slightly increased from 40% to 45%

for vehicle emissions, accompanied by a decrease of heterogeneous reactions on ground surface from 47% to 42% during the COVID-19 lockdown. This further highlights the importance of vehicle emissions to nocturnal HONO sources in Beijing.

In lines 628-640 in the revised manuscript, we added a sentence “To explore whether meteorological factors have an impact on the sources of HONO, we conducted the budget analysis of HONO using the deweathered pollutant concentrations. The results are shown in Fig. S10. When compared with the sources of HONO calculated using the raw concentration dataset (Fig. S9), it can be seen that deweathering has little effect on the daytime sources of HONO. For the nighttime source of HONO, however, deweathering caused the proportion of traffic emissions during BCNY increasing from 53% to 63% before the CNY or from 40% to 45% during the COVID-19 lockdown. The contribution of heterogeneous reactions of NO₂ on ground surfaces decreased from 31% to 19% before the CNY or from 47% to 42% during the COVID-19 lockdown. These results further highlight the importance of vehicle emissions to nocturnal HONO sources in Beijing.

Therefore, regardless of whether the impact of meteorological conditions on the source of HONO is considered, we can conclude that traffic-related emissions, rather than heterogeneous reactions of NO₂ were the main HONO source at night in Beijing in the typical emission patterns of air pollutants.”

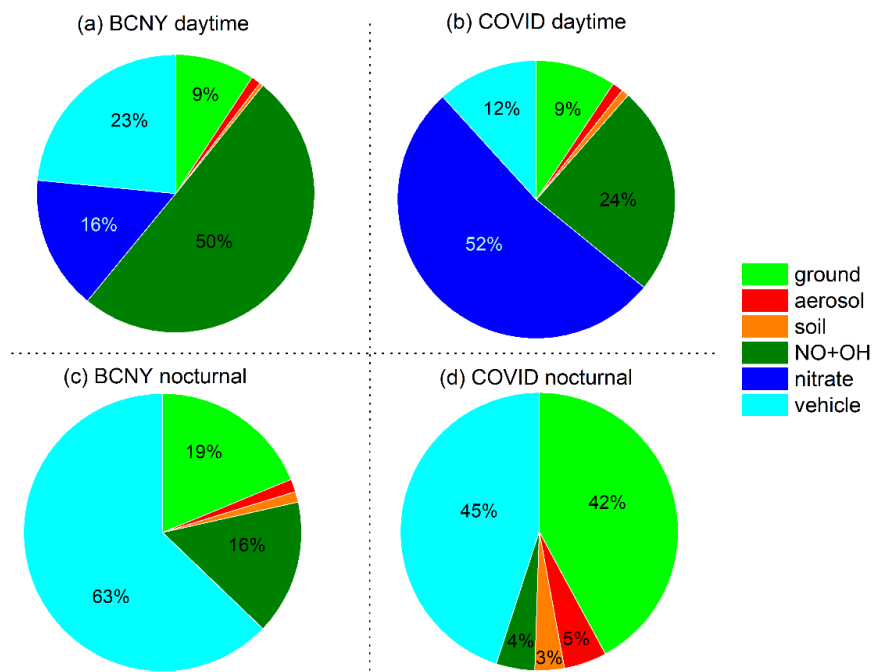


Figure R1. The percentage of daytime and nighttime contributions from different sources in (a,c) BCNY and (b,d) COVID. Pollutant concentrations are all de-weathering concentrations.

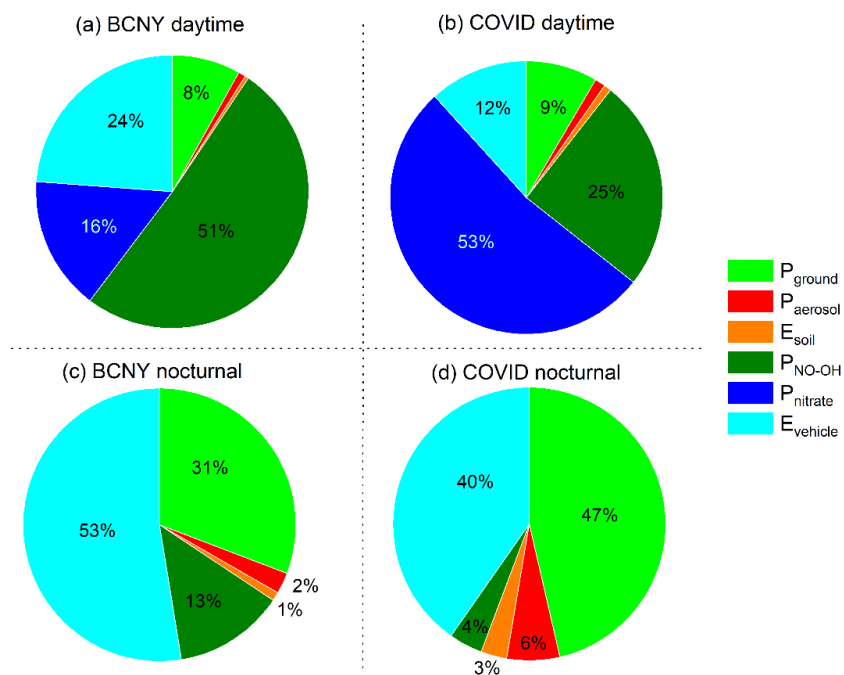


Figure R2. The percentage of daytime and nighttime contributions from different sources in (a,c) BCNY and (b,d) COVID. Pollutant concentrations are all raw concentrations.

2. The source function mapping for HONO (R2) is useful, it would be valuable to add to the manuscript conclusions the caveat that changes in air mass – as indicated on this figure (would be better to add the measurement site IAP also) exist between the two sampling periods, and this is a limitation of the analysis. I'm not sure how much can be drawn from the observation that there is an r of 0.79 and gradient of 0.6 between two separate measurement locations. These points – and the wider dependence on a large number of parameters, all inevitably with some uncertainty – do have consequences for the accuracy/precision of the final values presented, but I am comfortable with the language used in the abstract – the authors could consider reflecting this point at the start of the conclusions also.

Response: Thank you for your good suggestions. We conducted a potential source contribution function of HONO for the two observation sites, as shown in Figure R3 (or Figure S2) found that they are highly consistent. However, this still cannot rule out the influence of meteorological changes. In lines 683-693 in the revised manuscript, we added a sentence “We conducted a potential source contribution function (PSCF, Fig S2) analysis in different periods, i.e., BCNY and COVID, at the BUCT station and further compared the PSCF of HONO at BUCT station with that at the Institute Atmospheric Physics (IAP) station, which is around 8 km from BUCT station, from January 24, 2022, to January 31, 2022, when the data were available. The PSCF patterns were highly similar in different periods or locations. These results mean that the air mass should be consistent during the COVID-19 lockdown and BCNY and HONO should be evenly distributed in Beijing. Thus, the impact of meteorological changes on the accuracy of observations cannot be ruled out, which is also a limitation of this study, but its influence should be comparable between BCNY and the COVID lockdown. And the conclusions drawn based on the observations at BUCT should represent the situation in Beijing”. And in lines 650-651 in the revised manuscript, we added a sentence “It is worth noting that in addition to primary emissions, meteorological changes will also affect changes in atmospheric pollutant concentrations”. And in lines 666-668 in the revised manuscript, we also added a sentence “Although we have tried to assess the impact of meteorological factors quantitatively, this still carries some uncertainty. In particular, uncertainty is inevitable for the source assessment of

substances such as HONO that are affected by a large number of parameters”.

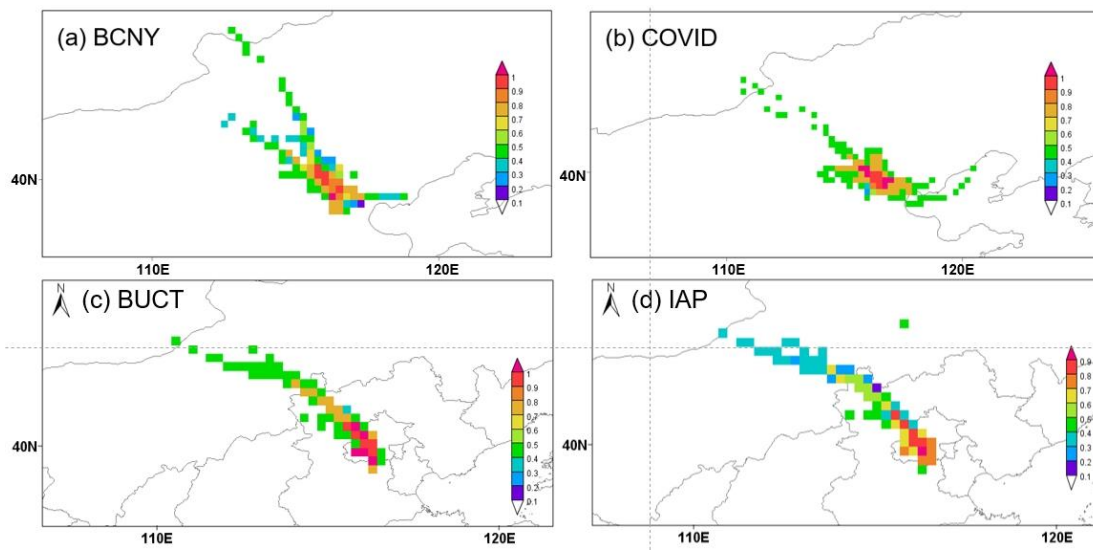


Figure R3. The potential source contribution function (PSCF) maps for the concentration of HONO (a and b are BCNY and COVID; c and d are BUCT and IAP stations, respectively). The comparison period of c and d is 2022.01.24-2022.01.31, and the trajectory of the air mass is 12 hours.

3. I'd ask the authors to consider how the statistical difference is presented – reviewing Figure S5 the ($\text{HONO}_{\text{corr}}/\text{NO}_2$) ratios / diurnal patterns clearly differ, but presenting these as the mean \pm SD values [(0.038 \pm 0.035), compared to a value of (0.042 \pm 0.034)] doesn't convey this to the audience nearly as well as the figure does – maybe include (the figure) in the main manuscript?

Response: Thank you for your good suggestions. When we present the values in Figure S5 as the average values for the whole day, the data cannot reflect the significant difference between the two periods. Therefore, we describe the average of the daytime with significant differences here to better highlight their statistical differences. In the revised manuscript in lines 478-480, we have modified it to “As shown in Fig. S5, the daytime peak of $\text{HONO}_{\text{corr}}/\text{NO}_2$ in P2 became more prominent compared with that in Fig. 3e, while the daytime (8:00 - 18:00) $\text{HONO}_{\text{corr}}/\text{NO}_2$ (0.022 ± 0.014) in P1 was significantly ($P < 0.05$) lower than that in P2 (0.040 ± 0.053)”.

4. Minor point - j_{NO_2} if derived from the MSM/Saunders et al approach doesn't account for clouds etc (it's a clear sky parameterization), but maybe this was normalised to measured j_{NO_2}

Response: Thank you for your suggestions. Our J_{NO_2} measurement equipment is a modified version of the original design by Junkermann et al., 1989 (Junkermann and Platt, 1989). The measuring principle is that the variation of the photolysis rate of NO_2 depends almost exclusively on the photochemical flux in the wavelength interval 300 to 420 nm. Photoelectric measurement of the photochemical flux in this spectral range gives a good approximation to the photolysis of NO_2 [$J_{(\text{NO}_2)}$]. The instrument consists of two detectors each covering one hemisphere with a nearly uniform angular response. The two detectors are mounted in opposite directions, one facing up towards the sky and one facing down. They consist of a set of quartz-domes, a set of optical filters, a radiation sensitive detector and an electronic board to provide a voltage output signal. Since the $J_{(\text{NO}_2)}$ photometer measures the radiant flux rather than directly measuring the photodecomposition rate of NO_2 , the instrument was calibrated to obtain standardized J_{NO_2} according to the photometer standards.

References:

JUNKERMANN, W. and PLATT, U.: A Photoelectric Detector for the Measurement of Photolysis Frequencies of Ozone and Other Atmospheric Molecules, *Journal of Atmospheric Chemistry*, 8, 203-227, 1989.