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Special Section:
The COVID-19 Pandemic: Linking Health, Society and Environment

Key Points:
- Observed PAN concentrations during COVID-19 in Beijing were significantly increased despite reduction in precursor emissions
- The chemical transport model simulations show that enhanced photochemistry caused the increase of PAN
- Abnormal weather conditions and nonlinear chemical feedback contributed to the enhanced photochemistry

Supporting Information:
- Supporting Information S1

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Markedly Enhanced Levels of Peroxyacetyl Nitrate (PAN) During COVID-19 in Beijing

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Abstract High levels of secondary air pollutants during COVID-19 in China have aroused great concern. In Beijing, measured daily mean peroxyacetyl nitrate (PAN) concentrations reached 4 ppb over the lockdown period (24 January to 15 February), whose averages were 2–3 times that before lockdown (1–23 January). The lockdown PAN levels also reached a high historical record based on our long-term measurements (2016–2019). Unlike ozone and $PM_{2.5}$, PAN formation depends on less complex photochemistry between NO_x and volatile organic compounds (VOCs), providing a novel approach to investigate the wintertime photochemistry during COVID-19. The GEOS-Chem simulations suggest a markedly enhanced photochemistry by a factor of 2 during the lockdown. Change of meteorology featuring with anomalous wind convergence under higher temperatures is the main reason for enhanced photochemical formation of PAN, while chemically nonlinear feedbacks also play a role. Our results suggest implementing targeted VOC emission controls in the context of increasing photochemical pollution over this complex polluted region.

Plain Language Summary Outbreaks of the COVID-19 pandemic caused immediate implementation of lockdown policy in China, which drastically decreased emissions of primary air pollutants. Peroxyacetyl nitrate (PAN), as an important photochemical product, is controlled by reactions between NO_x and volatile organic compounds (VOCs) that were reduced substantially due to the lockdown. However, observed PAN levels in Beijing during the lockdown were markedly enhanced and were even much higher than the concentrations during the same periods in 2016–2019. Modeling results prove that this increase in PAN is driven by enhanced photochemistry, resulting from anomalous wind convergence under higher temperature and enhanced radical level in response to NO_x reduction. Our results suggest the necessity of reducing VOC emissions in controlling photochemical pollution even in the wintertime over China.

1. Introduction

Outbreaks of the COVID-19 pandemic triggered immediate implementation of a lockdown policy in China and other countries, resulting in drastic decreases in emissions of air pollutants (Bauwens et al., 2020; Huang et al., 2020; Le et al., 2020; Sharma et al., 2020; Shi & Brasseur, 2020; Sicard et al., 2020). Satellite data indicated that nitrogen dioxide (NO_2) column concentrations over Chinese cities decreased by 40% due to lockdown measures (Bauwens et al., 2020). However, Huang et al. (2020) showed that $PM_{2.5}$ and ozone (O_3) concentrations in the North China Plain (NCP) surprisingly increased during the lockdown, and they attributed this to the enhanced secondary formation of air pollutants. However, the underlying reasons for accelerated atmospheric chemistry during the COVID-19 are still not fully explored.

Peroxyacetyl nitrate (PAN) is an important photochemical pollutant that is formed through reactions between NO_2 and peroxyacetyl radical (PA), which is usually thought to be photochemical product of volatile organic compounds (VOCs; Fischer et al., 2014; Singh et al., 1992; Xue et al., 2014). In this study, a high PAN level of 4 ppb in Beijing was observed during the COVID-19. As an indicator of active photochemistry, this provided an opportunity to investigate the photochemical level during this unique emission reduction period and facilitated our understanding of photochemical processes in wintertime.

PAN and O_3 are both photochemical pollutants, but they respond differently to their precursors and temperature. O_3 sensitivity to its precursors is more complicated than that of PAN and depends on whether O_3 production is under a NO_x-limited or VOC-limited regime (Jin & Holloway, 2015; Li et al., 2019; T. Wang et al., 2017). However, the relationship between PAN formation and its precursors is simple; that is, PAN concentration decreases with the reduction in VOCs and NO_x emissions (Fischer et al., 2014; Qiu, Ma, et al., 2019). PAN is easily decomposed under high temperatures (Seinfeld & Pandis, 2006), while O_3 production rate increases with temperature (Sillman & Samson, 1995). In a conventional view, the winter PAN levels are low due to weak solar radiation (G. Zhang et al., 2014), and in summer, observed PAN pollution events are usually accompanied by O_3 pollution episodes (B. Zhang et al., 2017). However, recent studies in China found that PAN concentrations on winter haze days could be as high as those observed in summer photochemical smog (Liu et al., 2018; Qiu, Ma, et al., 2019; G. Zhang, Xia, et al., 2020; B. Zhang et al., 2019), suggesting potential contributions from unfavorable meteorological conditions and local photochemical reactions (Qiu, Ma, et al., 2019; J. Zhang, Guo, et al., 2020).

In this study, we conducted observational and modeling analyses to explain the markedly enhanced PAN levels in Beijing during the lockdown and to explore the role of photochemistry in wintertime. First, this work shows the observed high PAN levels in Beijing during the lockdown, and unfavorable meteorological conditions over the same period are also displayed. Then, the impacts of changes in photochemistry will be examined using combined observations and simulations. In addition, we utilized sensitivity simulations to quantify the impacts of changes in emissions and meteorology on enhanced photochemistry during the lockdown period and to determine the driver of the observed high PAN levels. This study will help us better understand the increasingly important winter photochemistry in the NCP.

2. Methods

2.1. Observations

We conducted measurements of PAN at two sites in Beijing, that is, an urban site at Minzu University of China (39.95°N, 116.32°E) and a rural site at Shangdianzi regional Global Atmosphere Watch Station (40.65°N, 117.12°E). The detailed geophysical locations of the two sites are shown in Figure S1 in the supporting information. The urban site is located between the second and third ring roads in downtown Beijing, and it is mainly affected by anthropogenic sources. The rural site is located 100 km northeast of the urban center and near a small village. PAN concentrations were measured with an online gas chromatograph equipped with an electron capture detector (GC-ECD; Qiu, Lin, et al., 2019; Qiu, Ma, et al., 2019; Qiu et al., 2020). The time resolution was 5 min at both sites. Regular calibration checks were conducted every month to guarantee the quality of the PAN measurements. Observations at the urban and rural sites began in January 2019 and August 2015, respectively. We obtained PAN data from 1 January 2020 to 15 February 2020, as the data set of this study. The first 23 days (1–23 January), which were not affected by the lockdown policy, were considered as the preperiod, and the following 23 days (24 January to 15 February) were the lockdown period.

O_3, carbon monoxide (CO), and NO_2 concentrations were also simultaneously measured at the rural site using an O_3 analyzer (model 49i, Thermo Electron Corporation, USA), a CO analyzer (model 48i, Thermo Electron Corporation, USA), and a NO_2 analyzer (model 42i, Thermo Electron Corporation, USA), respectively. Considering the lack of measurements of chemical species concentrations at the urban site, we utilized the data at a site (Wanliu site) from the China Ministry of Ecology and Environment, which is located approximately 5 km north of the urban PAN measurement site.

2.2. GEOS-Chem Simulation

To investigate the cause of the observed high PAN levels during the lockdown period, we employed the GEOS-Chem chemical transport model (version 12.7.0; https://doi.org/10.5281/zenodo.3634864) driven by GEOS-FP assimilated meteorological data obtained from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). The GEOS-Chem model includes detailed ozone-NO_x-VOC-PM-halogen tropospheric chemistry (Li et al., 2019). The nested grid version of the

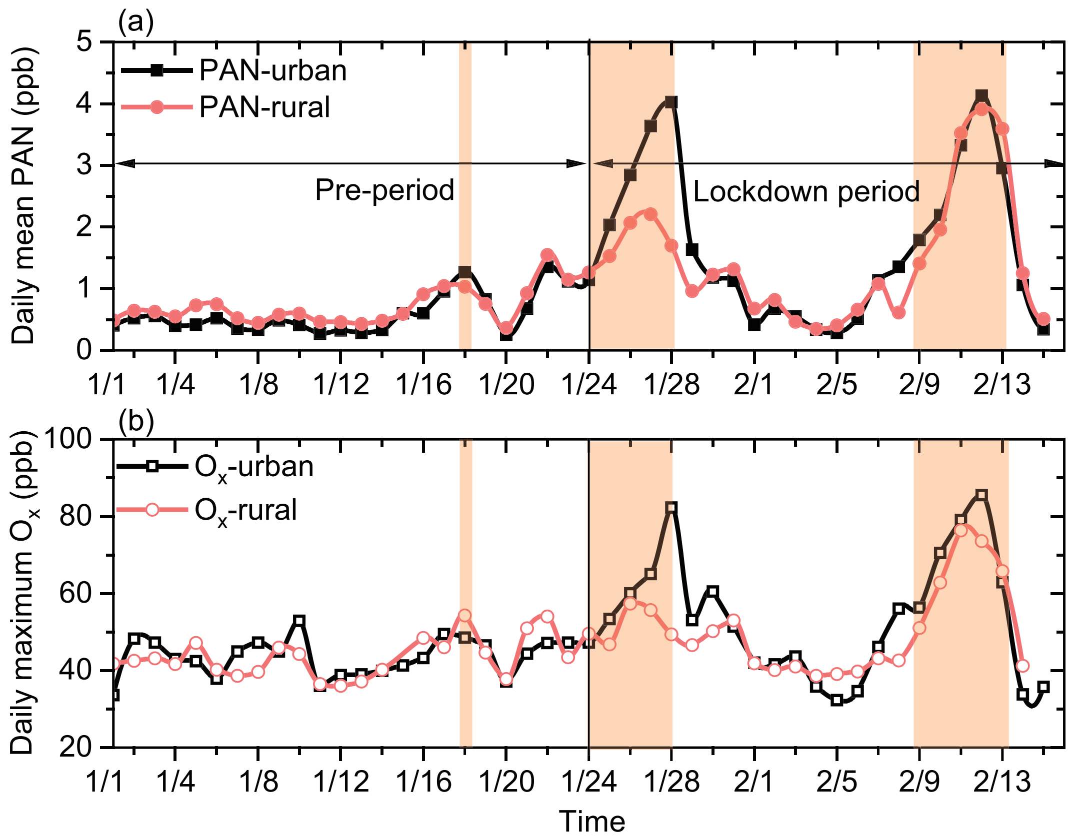

Figure 1. Observed (a) daily mean PAN and (b) daily maximum O_x concentrations (ppb) at the urban (black lines) and rural sites (red lines) in Beijing during January–February 2020. The orange shadings denote the days when daily mean $PM_{2.5}$ concentrations in Beijing exceed 75 μg m^{-3}.

model with a horizontal resolution of 0.25° × 0.3125° over eastern China (30–45°N, 108–125°E) was used. For anthropogenic emissions, we used the MEIC inventory (Zheng et al., 2018), which was developed by Tsinghua University (http://www.meicmodel.org/) and has been widely applied in simulating photochemical pollution (Gong & Liao, 2019; Hu et al., 2016; Lu et al., 2019) and haze events (Chen, Gao, et al., 2019; Chen, Zhu, et al., 2019; Qiu et al., 2017) in China.

To identify the impacts of emission reduction and changes in meteorology on PAN levels, we performed the following sensitivity simulations: (1) PRE, a simulation before the lockdown (23 days, from 1 to 23 January) with standard emissions; (2) LKD, a simulation during the lockdown period (23 days, from 24 January to 15 February) with an emission reduction scenario (decreased NO_x and VOC emissions by 60% and 30% over the model nested domain, respectively), and the detailed emission reduction method is shown in Table S1; and (3) LKD_Emis, which was the same as the LKD simulation, but the emission rates were those used in the PRE simulation. The contributions of emission reductions and changes in meteorology could be quantified by (LKD-LKD_Emis) and (LKD_Emis-PRE), respectively.

3. Results and Discussion

3.1. Observed High PAN Levels During the Lockdown Period

Figure 1 displays the observed time series of 24-hr mean PAN and daily maximum O_x ($O_x = O_3 + NO_2$) concentrations in Beijing during the preperiod (1–23 January) and lockdown period (24 January to 15 February). The observed PAN concentrations at the two sites during the preperiod were relatively low, with daily mean values of 0.26–1.55 ppb. However, PAN concentrations increased to 0.29–4.13 ppb at the two sites during the lockdown period, despite a substantial reduction in anthropogenic precursor emissions (Bauwens et al., 2020; Huang et al., 2020). Statistical results show that the mean PAN concentrations during the

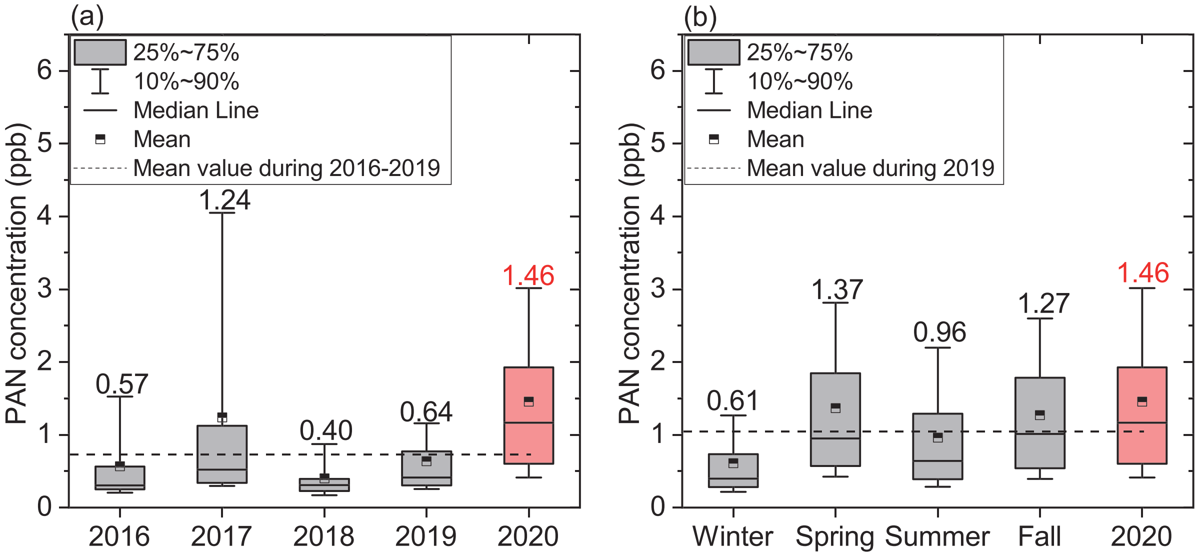

Figure 2. Box plots of (a) observed PAN concentrations (ppb) in Beijing during the lockdown period (24 January to 15 February) in 2020 (red) and those during the same periods in 2016–2019 (gray); (b) PAN concentrations (ppb) in Beijing during the lockdown period (24 January to 15 February) in 2020 (red) and seasonal mean concentrations in 2019 (gray). The dashed lines denote the average values during 2016–2019 (a) and during the four seasons in 2019 (b). The numbers on the top of the boxes denote the average values (ppb).

lockdown period at the urban (1.68 ppb) and rural sites (1.46 ppb) were 2.9 and 2.1 times the values during the preperiod, respectively. The O_x concentrations exhibited a similar increasing trend. The maximum values of daily maximum O_x concentrations at the urban and rural sites were 85.5 and 76.4 ppb, respectively, while the corresponding values during the preperiod were only 52.9 and 54.3 ppb. These results reveal that the concentrations of major photochemical pollutants in Beijing increased substantially during the lockdown period despite reductions in precursor emissions.

PAN concentrations during the lockdown period were also much higher than historical records. Figure 2a compares the PAN concentration in Beijing during the lockdown period with historical statistics, and here, we use the PAN concentration at the rural site where continuous observations over 2016–2020 were available. The PAN concentration during the lockdown period in 2020 (1.46 ppb) was 1.2–3.7 times that during the same periods in 2016–2019. We also compared PAN data with seasonal mean values in 2019 (Figure 2b). The results show that the PAN concentration during the lockdown period was 139%, 52%, 15%, and 7% higher than the mean values in winter, summer, fall, and spring, respectively. It should be noted that the concentration was even slightly higher than that in spring, when the maximum annual PAN concentration usually exhibited (Lee et al., 2013; Qiu et al., 2020). In addition, the PAN concentration in Beijing during the lockdown period was approximately 2–6 times that determined in previous observations during winter in Seoul (0.57–0.69 ppb; Lee et al., 2013) and Munich (0.24–0.27 ppb; Rappenglück et al., 1993), suggesting a high wintertime level of PAN over the NCP region. The O_x level during 2020 lockdown period is higher than those in the same period of 2017–2019 (Figure S2), also suggesting a relatively high photochemical pollutant level in spite of precursor reductions during lockdown.

3.2. Meteorological Conditions During the Lockdown Period

Southerly winds in the NCP region were previously reported to be favorable for the accumulation of PAN and its precursors in winter Beijing (Qiu, Ma, et al., 2019), and meteorology could play an important role in the observed PAN level. Thus, we first check meteorological conditions during the lockdown period in the following. As illustrated in Figure 1, there were two PAN pollution events (24–28 January and 9–13 February) with the co-occurrence of $PM_{2.5}$ pollution episodes during the lockdown period (Figure S3). The daily mean $PM_{2.5}$ concentrations at the urban site were 168 and 222 μg m^{-3} during these two events (Figure S3), when the corresponding PAN concentrations exceeded 4 ppb (Figure 1). The two haze events associated with high PAN levels suggest unfavorable meteorological conditions during the lockdown

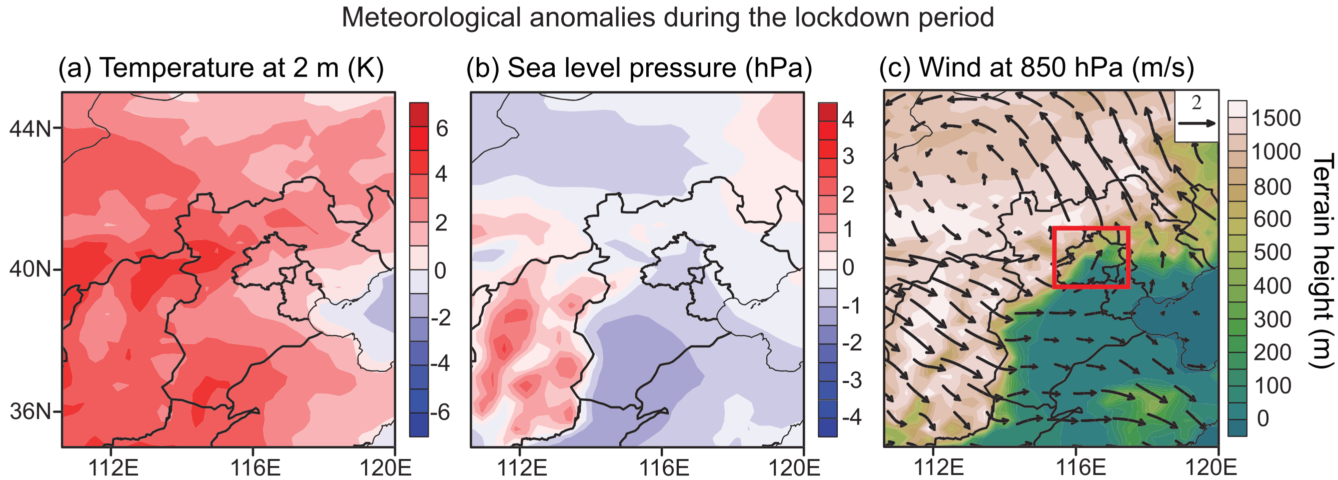

Figure 3. Differences in (a) temperature at 2 m (K), (b) sea level pressure (hPa), and (c) horizontal wind at 850 hPa (m s^{-1}) between the lockdown period (24 January to 15 February) and preperiod (1–23 January) over the NCP region. The terrain height (m, contour) and location of Beijing (red square) are also shown in (c).

period (Le et al., 2020). Figure 3 compares differences in temperature, sea level pressure, and horizontal winds at 850 hPa between the lockdown period and preperiod (LKD-PRE) over the NCP region from GEOS-FP reanalysis data. Surface temperature during the lockdown period increased by 1–3°C in Beijing and the middle part of the NCP. The increase in temperature during haze events has also been previously reported (Cai et al., 2017; Y. Wang et al., 2014). Relative to meteorological conditions during the preperiod, anomalous wind convergence at 850 hPa associated with lower sea level pressure during the lockdown period was found north of the NCP. As Beijing is surrounded by mountains on three sides (Figure S1), southerly anomalies are conducive to the accumulation of pollutants in the piedmont. Previous studies (Liu et al., 2018; Qiu, Ma, et al., 2019) have shown that PAN's lifetime could exceed several days in winter over the NCP region because it is hardly decomposed under low temperatures. Thus, it is of interest to determine whether the observed high PAN levels during the lockdown period were caused by PAN transport from the surrounding area or local enhanced photochemistry.

3.3. Enhanced Photochemistry During the Lockdown Period

To figure out the cause of enhanced PAN levels, we simulated PAN variations during January–February 2020 over northern China using the GEOS-Chem model. The model successfully captured PAN variations at urban and rural sites, with correlation coefficient (R) of 0.87 and 0.97, respectively, and the two pollution events were also well simulated (Figure S4). We conducted diagnosis analysis to classify the processes influencing PAN into convection, dry deposition, vertical mixing, transport, and chemistry in the boundary layer. As shown in Figure 4a, chemical reactions contribute to PAN formation, while transport induces a decrease in PAN due to relatively higher PAN concentrations in Beijing than in its surroundings. This means that the observed high PAN levels were attributed to local chemical formation instead of direct transport from other regions. The average PAN flux induced by chemical processes was 10.9 g s^{-1} during the lockdown period, which was 2.3 times that during the preperiod. Moreover, the temporal variation in the net PAN production rate (P[PAN]) also showed a significant increase from 1 January to 15 February (Figure 4b). On average, P[PAN] in Beijing during the lockdown period increased by 189% (Figure 4d), and an increase of 100–350% was also found in the middle and northeastern NCP region where high P[PAN] was simulated (Figure 4c). During the two pollution events, daily P[PAN] in Beijing increased by 130–600%, implying an important contribution from the two events. The higher P[PAN] during the lockdown period was also accompanied by an increase in major atmospheric oxidants (OH, NO_3, and O_x; Figure S5), suggesting the major role of enhanced photochemistry and atmospheric oxidation capacity during the emission reduction period. Moreover, the slopes of the linear fitting of PAN versus CO during the lockdown period were 4.5 and 2.2 times those during the preperiod at the urban and rural sites, respectively (Figure S6), which also indicates the likely enhanced local photochemistry during the lockdown from an observational perspective.

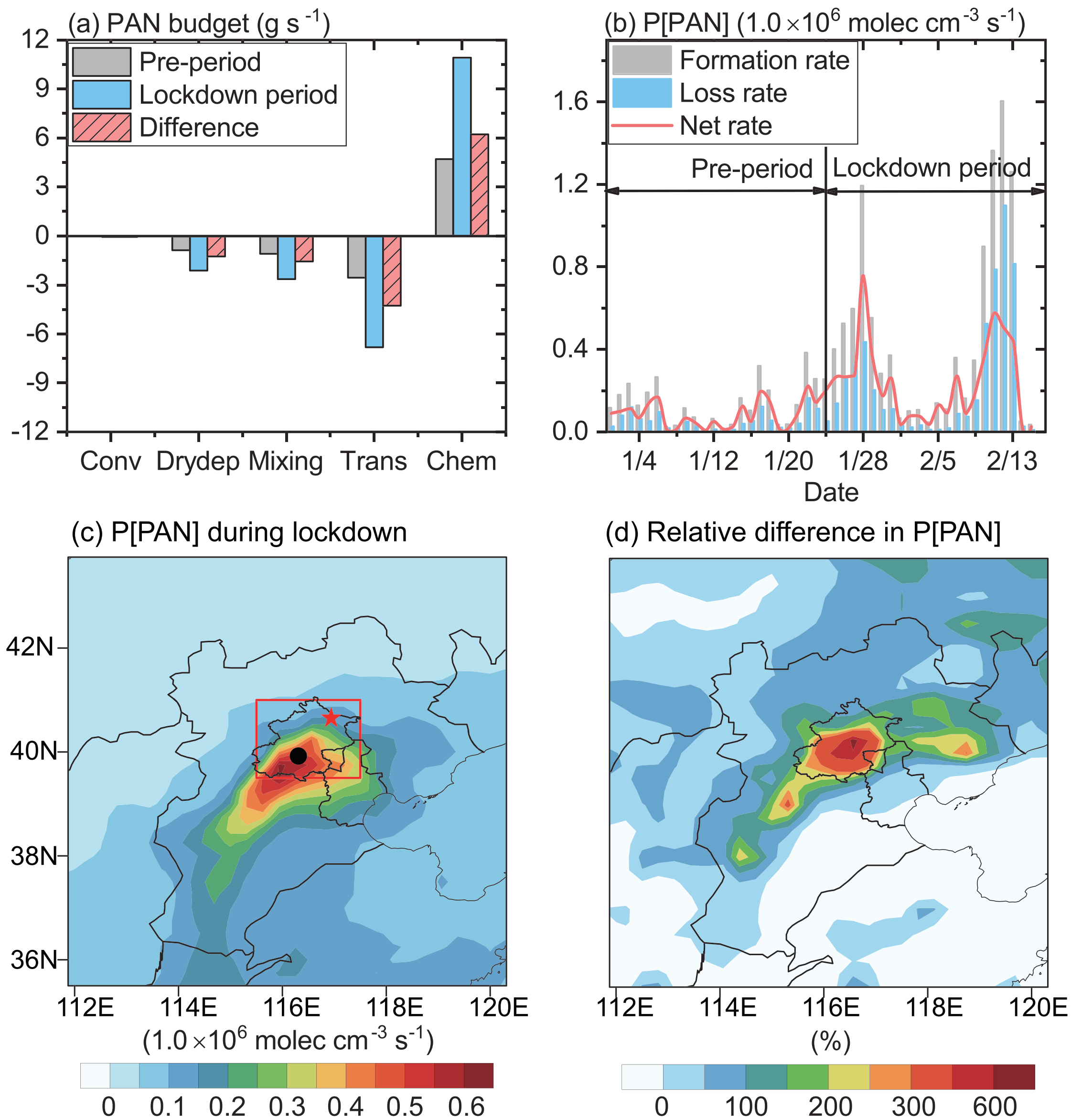

Figure 4. (a) Simulated daily mean PAN budget in the boundary layer (mass flux from each process, g s^{-1}) in Beijing during the lockdown period (24 January to 15 February) and preperiod (1–23 January), as well as the difference between them; the chemical and physical processes include convection (Conv), dry deposition (Drydep), vertical mixing (Mixing), transport (Trans), and chemistry (Chem). (b) Simulated daily PAN formation rate, loss rate, and net production rate (P[PAN], 1.0×10^6 molec cm^{-3} s^{-1}) in Beijing during the lockdown period and preperiod. (c) Simulated spatial distributions of P[PAN] (1.0×10^6 molec cm^{-3} s^{-1}) during the lockdown period over the NCP region. (d) Simulated spatial distributions of relative differences in P[PAN] (%) during the lockdown period and preperiod over the NCP region. The red square, black dot, and red star in Figure 4c denote the locations of average domain used in Figures 4a and 4b, urban and rural measurement sites, respectively.

A greater reduction in NO_x than VOCs in urban NCP could result in a weakened NO_x titration effect and enhanced free radical formation, accelerating photochemical reactions (T. Wang et al., 2017). On the other hand, increased precursor accumulation and temperature due to unfavorable meteorological conditions could also result in photochemical enhancement and thus an increase in P[PAN]. To determine the cause of enhanced photochemistry, we conducted sensitivity simulations using the GEOS-Chem model to examine the impacts of changes in meteorology and emission (Figure 5). The results demonstrated that the reduction in PAN precursors (LKD-LKD_Emis) could account for a 5–25% (average: 17%) decrease in the PAN concentration in Beijing (Figure 5b). Previous modeling study reported that PAN concentration generally responds more strongly to precursor emission changes (Fischer et al., 2014, their Figure 4), which is also consistent

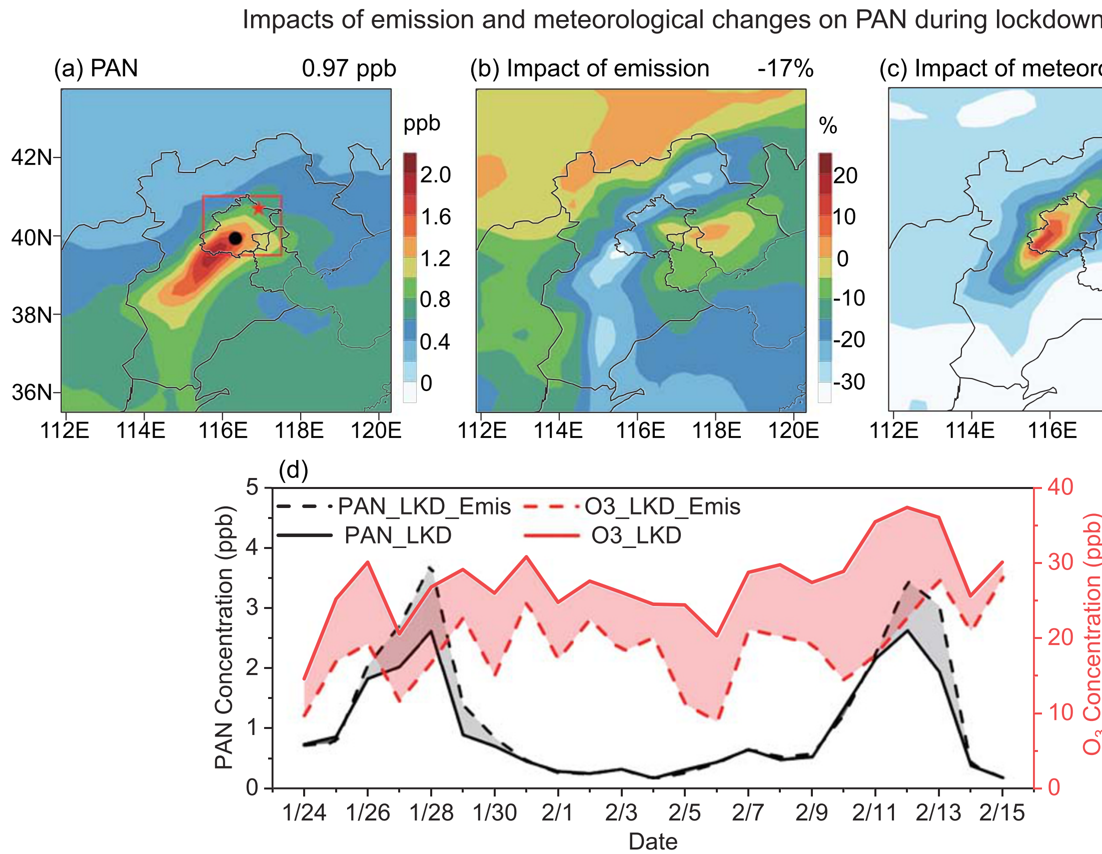

Figure 5. (a) Simulated PAN concentrations (LKD) during the lockdown period (24 January to 15 February) over the NCP region (ppb). (b) Simulated impacts of emission reduction on PAN concentrations ([LKD-LKD_Emis]/LKD_Emis) during the lockdown period over the NCP region (%). (c) Simulated impacts of changes in meteorology on PAN concentrations ([LKD_Emis-PRE]/PRE) during the lockdown period over the NCP region (%). The character in the upper right corner of each plot represents the average value in the red square. (d) Simulated daily concentrations of PAN (black lines) and O_3 (red lines) averaged over Beijing during the lockdown period (24 January to 15 February) in the LKD case (solid) and LKD_Emis (dash) case. The red square, black dot, and red star in Figure 5a denote the locations of average domain used in Figure 5d, urban and rural measurement sites, respectively.

with our previous results (Qiu, Ma, et al., 2019). In this study, the 17% decrease in PAN concentration was much lower than the lockdown emission reductions (60% in NO_x emissions and 30% in VOC emissions), implying that the offset effect from enhanced radicals and oxidants from ozone chemical nonlinearity originated from a greater reduction in NO_x. This is supported by Figure S7 which shows the simulated concentrations of OH, NO_3, and O_3 in Beijing during the lockdown increased by 12%, 76%, and 47%, respectively, in response to emission reduction alone.

Remarkably, changes in meteorological conditions before and after the lockdown period could lead to a 100–450% (average: 185%) increase in PAN concentration in Beijing during the lockdown period (Figure 5c), and the largest increase due to changes in meteorology appears in the area with a high PAN concentration (Figure 5a). As such, photochemical enhancement due to changes in meteorology was the main cause of the observed high PAN level in Beijing. On the one hand, precursor accumulation induced by anomalous southerly winds might contribute to the enhancement of P[PAN] during polluted days. Model results show that toluene and ethylene concentrations increased by approximately 50% and 125%, respectively, during polluted days due to unfavorable meteorology after 24 January, partially offsetting the effects of emission reduction. On the other hand, higher temperatures during the lockdown period than during the preperiod could induce higher OH and peroxyacetyl radical production rates, as the spatial pattern of increased OH radicals (Figure S5) during the lockdown period resembled the changes in temperature (Figure 3). This contributes to enhanced atmospheric oxidation capacity and high PAN levels.

Based on our analysis, enhanced photochemistry due to meteorological changes is the main cause of the observed high PAN levels in Beijing and surrounding areas in combination with observational and simulation results. The southerly anomalies facilitate precursor accumulation, and higher temperatures accelerated VOC oxidation, leading to enhanced photochemistry during the lockdown period. Further analysis shows that if emissions continued as business as usual (LKD_Emis case), the average PAN concentration in Beijing during lockdown polluted days (24–28 January and 9–13 February) increased by 0.39 ppb (40%) in comparison with LKD case (Figure 5d), implying the necessity for more strict reductions in precursor emissions to control photochemical and secondary pollution under unfavorable meteorological conditions. In contrast, the O_3 concentration with standard emission scenario was reduced by 32% (Figure 5d) due to nonlinear chemical feedbacks, which is similar to previous studies (Huang et al., 2020; Le et al., 2020). These suggest the government paying more attention to decrease VOC emissions so that the impacts of chemical nonlinear effects could be minimized. Moreover, as suggested by Qiu, Ma, et al. (2019), the transport of PAN precursors from the polluted NCP region has a great impact on the local PAN concentration in Beijing. Therefore, interregional prevention and control of precursor emissions is necessary in reducing photochemical pollution in Beijing and the whole NCP region.

4. Conclusions

In this study, we performed integrated observational and modeling analysis to explain the increased concentrations of photochemical pollutant PAN in Beijing during the 2020 lockdown period (24 January to 15 February). The observed PAN concentration in Beijing during the lockdown reached the high level of 4 ppb, and the averages were approximately 2–3 times that before lockdown (1–23 January). These values were 1.2–3.7 times those in the same periods of 2016–2019. The GEOS-Chem simulations suggested that the observed high PAN levels during the lockdown were attributed to enhanced local photochemistry. Further model analysis revealed that the abnormal meteorological conditions (i.e., a 1–3°C increase in temperature and anomalous wind convergence) led to precursor accumulation and accelerated VOC oxidation, contributing most to the enhanced photochemistry.

In addition, chemically nonlinear feedback also played a role in enhanced photochemistry, because of the much smaller decrease of PAN concentration (17%) in response to sharp precursor reduction (i.e., 60% in NO_x emissions and 30% in VOC emissions). Sensitivity simulations show that if emissions continued as business as usual, PAN concentrations in Beijing during polluted days could increase by 0.39 ppb (40%). Our results suggest the necessity of reducing more VOC emissions in controlling photochemical pollution under unfavorable meteorological conditions over the complex polluted NCP region. In this study, the markedly enhanced PAN levels during COVID-19 from our long-term PAN records demonstrate that PAN is an effective indicator of wintertime photochemistry. Considering the possibly important role of photochemical pollutants on $PM_{2.5}$ formation, further studies targeting the understanding of wintertime photochemical pollutants (e.g., O_3 and PAN) are urgently needed. PAN is not measured as criteria pollutants, and we call for more efforts to improve the ability of photochemical monitoring in China.

Conflict of Interest

The authors declared that they have no conflicts of interest related to this work.

Data Availability Statement

The GEOS-FP assimilated meteorological data are obtained from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO) (https://gmao.gsfc.nasa.gov/GMAO_products/NRT_products.php). The model output and PAN observations used in this study can be accessed via doi (https://doi.org/10.7910/DVN/LUX8JX).

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References

Bauwens, M., Compernolle, S., Stavrakou, T., Müller, J. F., Gent, J., Eskes, H., et al. (2020). Impact of coronavirus outbreak on NO_2 pollution assessed using TROPOMI and OMI observations. *Geophysical Research Letters*, *47*, e2020GL087978. https://doi.org/10.1029/2020GL087978

Cai, W., Li, K., Liao, H., Wang, H., & Wu, L. (2017). Weather conditions conducive to Beijing severe haze more frequent under climate change. *Nature Climate Change*, *7*(4), 257–262. https://doi.org/10.1038/nclimate3249

Chen, L., Gao, Y., Zhang, M., Fu, J. S., Zhu, J., Liao, H., et al. (2019). MICS-Asia III: Mult-model comparison and evaluation of aerosol over East Asia. *Atmospheric Chemistry and Physics*, *19*(18), 11,911–11,937. https://doi.org/10.5194/acp-19-11911-2019

Chen, L., Zhu, J., Liao, H., Gao, Y., Qiu, Y., Zhang, M., et al. (2019). Assessing the formation and evolution mechanisms of severe haze pollution in Beijing-Tianjin-Hebei region by using process analysis. *Atmospheric Chemistry and Physics*, *19*(16), 10,845–10,864. https://doi.org/10.5194/acp-19-10845-2019

Fischer, E. V., Jacob, D. J., Yantosca, R. M., Sulprizio, M. P., & Millet, D. B. (2014). Atmospheric peroxyacetyl nitrate (PAN): A global budget and source attribution. *Atmospheric Chemistry and Physics*, *14*(5), 2679–2698. https://doi.org/10.5194/acp-14-2679-2014

Gong, C., & Liao, H. (2019). A typical weather pattern for the ozone pollution events in North China. *Atmospheric Chemistry and Physics*, *19*(22), 13,725–13,740. https://doi.org/10.5194/acp-19-13725-2019

Hu, J., Chen, J., Ying, Q., & Zhang, H. (2016). One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system. *Atmospheric Chemistry and Physics*, *16*(16), 10,333–10,350. https://doi.org/10.5194/acp-16-10333-2016

Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., et al. (2020). Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China. *National Science Review*. https://doi.org/10.1093/nsr/nwaa137

Jin, X., & Holloway, T. (2015). Spatial and temporal variability of ozone sensitivity over China observed from the ozone monitoring instrument. *Journal of Geophysical Research: Atmospheres*, *120*, 7229–7246. https://doi.org/10.1002/2015JD023250

Le, T., Wang, Y., Liu, L., Yang, J., Yung, Y. L., Li, G., & Seinfeld, J. H. (2020). Unexpected air pollution with marked emission reductions during the COVID-19 outbreak in China. *Science*, *369*(6504), 702–706. https://doi.org/10.1126/science.abb7431

Lee, J. B., Yoon, J. S., Jung, K., Eom, S. W., Chae, Y. Z., Cho, S. J., et al. (2013). Peroxyacetyl nitrate (PAN) in the urban atmosphere. *Chemosphere*, *93*(9), 1796–1803. https://doi.org/10.1016/j.chemosphere.2013.06.019

Li, K., Jacob, D. J., Liao, H., Zhu, J., Shah, V., Shen, L., et al. (2019). A two-pollutant strategy for improving ozone and particulate matter air quality in China. *Nature Geoscience*, *12*(11), 906–910. https://doi.org/10.1038/s41561-019-0464-x

Liu, L., Wang, X., Chen, J., Xue, L., Wang, W., Wen, L., et al. (2018). Understanding unusually high levels of peroxyacetyl nitrate (PAN) in winter in Urban Jinan, China. *Journal of Environmental Sciences*, *71*, 249–260.

Lu, X., Zhang, L., Chen, Y., Zhou, M., Zheng, B., Li, K., et al. (2019). Exploring 2016–2017 surface ozone pollution over China: Source contributions and meteorological influences. *Atmospheric Chemistry and Physics*, *19*(12), 8339–8361. https://doi.org/10.5194/acp-19-8339-2019

Qiu, Y., Liao, H., Zhang, R., & Hu, J. (2017). Simulated impacts of direct radiative effects of scattering and absorbing aerosols on surface-layer aerosol concentrations in China during a heavily polluted event in February 2014. *Journal of Geophysical Research: Atmospheres*, *122*, 5955–5975. https://doi.org/10.1002/2016JD026309

Qiu, Y., Lin, W., Li, K., Chen, L., Yao, Q., Tang, Y., & Ma, Z. (2019). Vertical characteristics of peroxyacetyl nitrate (PAN) from a 250-m tower in northern China during September 2018. *Atmospheric Environment*, *213*, 55–63. https://doi.org/10.1016/j.atmosenv.2019.05.066

Qiu, Y., Ma, Z., & Li, K. (2019). A modeling study of the peroxyacetyl nitrate (PAN) during a wintertime haze event in Beijing, China. *Science of Total Environment*, *650*(Pt 2), 1944–1953. https://doi.org/10.1016/j.scitotenv.2018.09.253

Qiu, Y., Ma, Z., Lin, W., Quan, W., Pu, W., Li, Y., et al. (2020). A study of peroxyacetyl nitrate at a rural site in Beijing based on continuous observations from 2015 to 2019 and the WRF-Chem model. *Frontiers of Environmental Science & Engineering*, *14*(4), 71.

Rappenglück, B., Kourtidis, K., & Fabian, P. (1993). Measurements of ozone and peroxyacetyl nitrate (PAN) in Munich. *Atmospheric Environment*, *27*(3), 293–305.

Seinfeld, J. H., & Pandis, S. N. (2006). *Atmospheric chemistry and physics: From air pollution to climate change*. Hoboken, New Jersey, USA.

Sharma, S., Zhang, M., Anshika, G. J., Zhang, H., & Kota, S. H. (2020). Effect of restricted emissions during COVID-19 on air quality in India. *Science of the Total Environment*, *728*, 138878. https://doi.org/10.1016/j.scitotenv.2020.138878

Shi, X., & Brasseur, G. P. (2020). The response in air quality to the reduction of Chinese economic activities during the COVID-19 outbreak. *Geophysical Research Letters*, *47*, e2020GL088070. https://doi.org/10.1029/2020GL088070

Sicard, P., Marco, A. D., Agathokleous, E., Feng, Z., Xu, X., Paoletti, E., et al. (2020). Amplified ozone pollution in cities during the COVID-19 lockdown. *Science of Total Environment*, *735*, 139542. https://doi.org/10.1016/j.scitotenv.2020.139542

Sillman, S., & Samson, P. J. (1995). Impacts of temperature on oxidant photochemistry in urban, polluted rural and remote environments. *Journal of Geophysical Research*, *100*(D6), 11,497–11,508. https://doi.org/10.1029/94JD02146

Singh, H. B., O'Hara, D., Herlth, D., Bradshaw, D., Sandholm, S. T., Gregory, G. L., et al. (1992). Atmospheric measurements of peroxyacetyl nitrate and other organic nitrates at high latitudes: Possible sources and sinks. *Journal of Geophysical Research*, *97*(D15), 16,511–16,522. https://doi.org/10.1029/91JD00889

Wang, T., Xue, L., Brimblecombe, P., Lam, Y. F., Li, L., & Zhang, L. (2017). Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects. *Science of Total Environment*, *575*, 1582–1596. https://doi.org/10.1016/j.scitotenv.2016.10.081

Wang, Y. S., Yao, L., Wang, L. L., Liu, Z. R., Ji, D. S., Tang, G. Q., et al. (2014). Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China. *Science China Earth Sciences*, *57*(1), 14–25. https://doi.org/10.1007/s11430-013-4773-4

Xue, L., Wang, T., Wang, X., Blake, D., Gao, J., Nie, W., et al. (2014). On the use of an explicit chemical mechanism to dissect peroxy acetyl nitrate formation. *Environmental Pollution*, *195*(36), 39–47. https://doi.org/10.1016/j.envpol.2014.08.005

Zhang, B., Zhao, B., Zuo, P., Huang, Z., & Zhang, J. (2017). Ambient peroxyacyl nitrate concentration and regional transportation in Beijing. *Atmospheric Environment*, *166*, 543–550. https://doi.org/10.1016/j.atmosenv.2017.07.053

Zhang, B., Zhao, X., & Zhang, J. (2019). Characteristics of peroxyacetyl nitrate pollution during a 2015 winter haze episode in Beijing. *Environmental Pollution*, *244*, 379–387. https://doi.org/10.1016/j.envpol.2018.10.078

Zhang, G., Mu, Y., Liu, J., Zhang, C., Zhang, Y., Zhang, Y., & Zhang, H. (2014). Seasonal and diurnal variations of atmospheric peroxyacetyl nitrate, peroxypropionyl nitrate, and carbon tetrachloride in Beijing. *Journal of Environmental Sciences*, *26*(1), 65–74. https://doi.org/10.1016/S1001-0742(13)60382-4

Zhang, G., Xia, L., Zang, K., Xu, W., Zhang, F., Liang, L., et al. (2020). The abundance and inter-relationship of atmospheric peroxyacetyl nitrate (PAN), peroxypropionyl nitrate (PPN), O_3, and NO_y during the wintertime in Beijing, China. *Science of the Total Environment*, *718*, 137388. https://doi.org/10.1016/j.scitotenv.2020.137388

Zhang, J., Guo, Y., Qu, Y., Chen, Y., Yu, R., Xue, C., et al. (2020). Effect of potential HONO sources on peroxyacetyl nitrate (PAN) formation in eastern China in winter. *Journal of Environmental Sciences*, *94*, 81–87. https://doi.org/10.1016/j.jes.2020.03.039

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., et al. (2018). Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions. *Atmospheric Chemistry and Physics*, *18*(19), 14,095–14,111. https://doi.org/10.5194/acp-18-14095-2018

References From the Supporting Information

Zhang, R., Zhang, Y., Lin, H., Feng, X., Fu, T. M., & Wang, Y. (2020). NO_x emission reduction and recovery during COVID-19 in East China. *Atmosphere*, *11*(4), 433. https://doi.org/10.3390/atmos11040433