Source–Receptor Relationship Revealed by the Halted Traffic and Aggravated Haze in Beijing during the COVID-19 Lockdown

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ABSTRACT: The COVID-19 outbreak greatly limited human activities and reduced primary emissions particularly from urban on-road vehicles but coincided with Beijing experiencing “pandemic haze,” raising the public concerns about the effectiveness of imposed traffic policies to improve the air quality. This paper explores the relationship between local vehicle emissions and the winter haze in Beijing before and during the COVID-19 lockdown based on an integrated analysis framework, which combines a real-time on-road emission inventory, in situ air quality observations, and a localized numerical modeling system. We found that traffic emissions decreased substantially during the COVID-19 pandemic, but its imbalanced emission abatement of NOx (76%, 125.3 Mg/day) and volatile organic compounds (VOCs, 53%, 52.9 Mg/day) led to a significant rise of atmospheric oxidants in urban areas, resulting in a modest increase in secondary aerosols due to inadequate precursors, which still offset reduced primary emissions. Moreover, the enhanced oxidizing capacity in the surrounding regions greatly increased the secondary particles with relatively abundant precursors, which was transported into Beijing and mainly responsible for the aggravated haze pollution. We recommend that mitigation policies should focus on accelerating VOC emission reduction and synchronously controlling regional sources to release the benefits of local traffic emission control.

INTRODUCTION

The unexpected COVID-19 pandemic in 2020, which coincided with the Spring Festival, the most important holiday in China, put the Chinese economy into a rapid stall. The Spring Festival migration reduced significantly the population in Beijing, with an estimated 39% decrease from the 22 million residents in normal times, while the Coronavirus pandemic lockdown further reduced human activities. The stay-at-home orders were initially imposed in Wuhan on January 23, 2020, one day before the eve of the Spring Festival. Soon afterward, the lockdown was applied to the whole country. In addition, a two-week compulsory quarantine was implemented for travelers to Beijing. The Spring Festival holiday and the Coronavirus restrictions led to widespread shutdowns and a near-halt in normal life and economic activities in Beijing and its surrounding cities. Because the lockdown policy significantly affected various human activities, its impacts on the anthropogenic emissions and air quality probably exceeded the effects of the well-known “activity-limited” events in the past, such as the US copper smelter strike or the Olympic Games in Beijing. Generally, pandemic lockdowns led to clearer skies in China and other places in the world. However, severe air pollution episodes occurred in Beijing during “the most silent spring,” leading to query the response of air pollution to anthropogenic activities.

The unexpected heavy pollution cast doubt on the current understanding of the source–receptor relationship in Beijing. Following scientific instructions, control measurements were undertaken over the past 5 years to reduce the sources of aerosol pollution. As a result, the PM2.5 annual concentration in Beijing decreased from 89.5 μg/m3 in 2013 to 42.0 μg/m3 in 2019, and heavy pollution days were also reduced from 58 days in 2013 to 4 days in 2019, providing confidence in source–receptor mechanisms supporting pollution control strategies. However, during the COVID-19 lockdown, the air quality index (AQI) frequently hit extremely unhealthy levels in Beijing, including January 25–29 and February 9–13, with a peak daily PM2.5 concentration reaching 218.3 μg/m3 on February 12, which is more than eight times the World Health Organization’s recommended level of 25 μg/m3 for 24 h average concentrations. Here, we show that the severe haze pollution, which occurred in spite of the considerable decrease...
of human activities, highlights weaknesses in our current source–receptor understanding.

The “pandemic haze” in Beijing raised great attention from the public and the government on the role of vehicle emissions regarding air pollution. Historically, vehicle emission control has been used as an effective way to relieve air pollution in megacities.9,10 Beijing undertook a lot of effort to reduce its traffic emissions through imposing strict control strategies on new vehicle registration, limiting car usage based on plate numbers, upgrading vehicle emission standards, and shifting to a greener transportation.11,12 In Beijing, a car license plate is regarded as a limited public resource. To get a conventional gasoline car, residents need to participate in a bimonthly lottery pool, competing with more than 3 million fellow residents with odds of around 1/2000. Beijing residents believed that these restrictions could help improve the air quality, as official reports showed that the vehicles contributed 45% to ambient PM$_{2.5}$ from local sources in Beijing.13 However, recent air pollution episodes in Beijing have raised several questions about the validity of this relationship. Although several recent studies have investigated the causes of the pandemic haze in China on a large scale, little attention has been paid to the detailed variations of vehicle emissions and its impacts on the aggravated haze formation in a megacity.14,15 What was the role of vehicular emission reductions in PM$_{2.5}$ pollution during the COVID-19 outbreak? Is traffic emission control still a necessary and effective way to relieve the winter haze in a megacity like Beijing?

Throughout this paper, we presented a source–receptor analysis on the COVID-19 “pandemic haze” events in Beijing based on emission inventory, air quality observations, and numerical models. Our study integrated multiple real-time traffic data around the COVID-19 outbreak and developed a novel realistic traffic emission inventory for Beijing. It was applied in a series of counterfactual modeling experiments by a localized chemical transport modeling system and a tracing-based source apportionment. Our aim is to understand the mechanisms and the role of local vehicle emission reductions for the “pandemic haze” and to propose the future development of vehicle emission control strategies.

## MATERIALS AND METHODS

### Emission Estimation Methods

In this work, we built an integrated analysis framework to investigate the role of vehicle emissions in winter haze pollution in Beijing around the COVID-19 outbreak and Spring Festival (Figure S1). First, we developed a street-level on-road vehicle emission (SLOVE) model to estimate the hourly vehicle emissions from both exhausts and evaporation in urban areas of Beijing. This model consists of two dynamic databases, including (a) hourly road speed and (b) the observed meteorological condition, and three static local traffic information databases, including (a) fleet composition, (b) road basic information, and (c) vehicle emission factors. The real-time traffic condition data through the application programming interface (API) to AMap (www.amap.com) was obtained to calculate the traffic flow based on the fitting single-regime models (Figure S2). The vehicle fleet composition was different for various road types, which was collected by the Beijing Municipal Commission of Transport (BMCT) based on the video data in typical roads (Figure S3). The basic emission factors for various species were from the guide book for on-road emissions published by the Ministry of Ecology and Environment of the People’s Republic of China (MEE)16 and corrected by the traffic conditions using the computer programme to calculate emissions from road transport (COPERT) model and the environmental condition using the Motor Vehicle Emission Simulator (MOVES)17,18 model. Detailed descriptions about this model are discussed in the Supporting Information section S1. In addition, the emissions from heavy duty trucks (HDTs) were evaluated based on a more accurate TrackATruck model developed in our previous research, which is driven by big data (trajectory signals of each HDT from the BeiDou Navigation Satellite System) with advantages of considering individual truck differences.19

In addition to the on-road emissions calculated in this study, emissions from other sectors applied in the numerical modeling were assembled from several recent studies to improve the precision of the emission inventory. It includes the urban anthropogenic emissions for Beijing, Tianjin, and the surrounding 26 major cities in northern China collected from an air pollution prevention plan proposed by the government (referred to “2 + 26” plan), shipping emissions from our previous research,20 other anthropogenic emissions in China from the Multi-resolution Emission Inventory for China (MEIC) model,21 and others listed in Table S1. The emission changes of other anthropogenic sources affected by COVID-19 were calculated, respectively (Table S2), based on the changes in the related industrial and residential activities (for details, see Supporting Information section S2).

### Air Quality and Meteorological Monitoring Data Sets

Measurements of air pollutants were analyzed to identify the changes in the characteristics of air quality due to the extreme COVID-19 lockdown in Beijing. The hourly air quality data at 34 stations covering most urban and rural areas of Beijing, including AQI, PM$_{2.5}$, and gaseous air pollutant (NO$_2$, O$_3$, CO, and SO$_2$) concentrations mainly based on Thermo Scientific samplers and analyzers, were obtained from the Beijing Municipal Environmental Monitoring Center (http://zx.bjmemc.com.cn/). The AQI was a comprehensive measure of air pollution: the index was determined by the maximum concentration of different conventional air pollutants, with a higher AQI meaning worse air quality. Hourly concentrations of five dominant chemical components of PM$_{2.5}$ including element carbon (EC), organic matter (OM), sulfate (SO$_4^{2-}$), nitrate (NO$_3$), and ammonium (NH$_4$) at three monitoring sites were collected from the National Research Program for Key Issues in Air Pollution Control. The ground-observed weather data obtained from the National Climate Data Center (ftp://ftp.ncdc.noaa.gov/pub/data/noaa/)—integrated surface database were used to correct the vehicle emission factors in the SLOVE model and to estimate the accuracy of meteorological predictions in numerical modeling. The geographic locations of these observation sites were marked in Figure S4.

### Configurations and Evaluations of Numerical Modeling

The Weather Research and Forecasting (WRF, version 3.8.1)—Community Multiscale Air Quality (CMAQ, version 5.2) model was applied to simulate the air quality in Beijing from January 10 to February 15 in 2020.22,23 The modeling system drew on the 4-nested run with a horizontal resolution at 1.33 km of the innermost domain, where an urban canyon model (UCM) with updated land use data and urban canyon parameters (UCPs) were applied in the WRF model to improve the prediction of the meteorological field (Figures S4 and S5).24 To reproduce the polluted days, the heterogeneous
reactions of SO$_2$ were incorporated into the CMAQ model to enhance the sulfate formation at a high relative humidity (RH). In addition, the CMAQ model (version 5.0.2) with the Integrated Source Apportionment Method (ISAM) was used to determine the source contributions of local sources (on-road vehicles, industry, domestic, and others) and regional sources to the PM$_{2.5}$ and its species concentrations in Beijing around the COVID-19 outbreak.26 Emissions, meteorological fields, and configurations remained the same in both CMAQ versions. Detailed model configurations are described in the Supporting Information section S3.

We evaluated the simulated NO$_2$, O$_3$, and PM$_{2.5}$ concentrations against ground-based observations (Table S3). The CMAQv5.2 model well captured the variations of air quality with correlation coefficients higher than 0.5 for all species. The overall model performance in predicting PM$_{2.5}$ was also within the recommended benchmarks (mean fractional bias (MFB) $\leq$ ±60% and mean fractional error (MFE) $\leq$ ±75%)77 and in line with other recent modeling studies in Beijing.18,19 Moreover, the performance of CMAQv5.2 in predicting major PM$_{2.5}$ chemical components is examined in detail in Figures S6 and S7 (statistics in Table S3). An acceptable performance was obtained with a small mean bias (MB) for each species, ranging from 0.3 to 4.3 $\mu$g/m$^3$. Although they were slightly overestimated with a mean bias (MB) for each species, ranging from 0.3 to 4.3 $\mu$g/m$^3$. An acceptable performance was obtained with a small normalized mean bias (NMB) ranging from 16 to 32%, especially on several clean days (January 21−22), we focused on the differences in air quality before and after this period. Because EC, mainly from primary emissions, was overpredicted simultaneously with the secondary components, the uncertainties in predicting meteorological fields were probably responsible for these biases. On some polluted days (e.g., January 18 and 27), the revised CMAQ model overestimated particulate sulfate and ammonium concentrations, which was partly attributed to the selection of uptake coefficients in a heterogeneous reaction of SO$_2$. Most recently, in-particle nitrate photolysis in heterogeneous oxidation of SO$_2$ has been proved to explain some differences between predicted and measured sulfate during winter haze,31,32 which can be applied in future modeling research to improve the predicted sulfate trend during the haze events. The performance of CMAQv5.0.2 in simulating EC and secondary inorganic aerosols was also reliable with MBs ranging from 0.3 to 3.6 $\mu$g/m$^3$. In contrast, owing to the missing secondary organic matter (SOM) formation pathways included in CMAQv5.2 (e.g., aging of semi-/intermediate-volatile organic compounds (S/IVOCs) and primary organic aerosols based on a volatility basis set (VBS) framework),33,34 the OM was underpredicted with an NMB of −63%. However, the underestimated SOM in CMAQv5.0.2 would not affect the source apportionment results because the SOM formation was not traced in the ISAM due to the existing limitation.26 The rigorous comparison of modeling results from CMAQv5.2, CMAQv5.0.1-ISAM, and field measurements provided confidence in the source contribution described in the results. Evaluation of model performance in predicting meteorological fields is described in the Supporting Information section S4.

Modeling Scenarios. To quantify the influence of vehicle emission reduction on air quality before and during the COVID-19 outbreak, we set up a series of scenarios in the WRF-CMAQ modeling system with different fluctuations in on-road emissions only, while other configurations remained the same as the BASE scenario, in order to eliminate the influence of meteorology and emissions from other sectors. The BASE scenario simulated the air quality changes with real emission variations. The S1 scenario was implemented for the COVID-19 lockdown period during which vehicle emissions were assumed to remain at their usual level similar to the ones before the pandemic without any reduction, while the nonvehicle emissions experienced the real reduction. The S2−S6 scenarios were conducted for the prelockdown period during which the assumed relative reduction of vehicle emissions changed from 0 to 100%, while emissions from nonvehicle sectors remained at their usual level without any reduction. The assumed vehicle emission reduction in the S2 scenario was the same as the real reduction due to the COVID-
19 outbreak, and it reached 100% in the S3 scenario. In S4−S6 scenarios, the vehicle NOx emission was assumed to be reduced by 100, 50, and 0% in the case of 100% hydrocarbon (HC) emission reduction from vehicles, respectively, while its PM2.5 emissions remained unchanged. Details on the setting and purpose of each scenario in numerical modeling are provided in Table S4.

RESULTS

Traffic Activity Variations and Emission Changes during the Lockdown. The overall research period was divided into three subperiods, all occurring in the early spring: the prelockdown period (referred to Pre, before January 20), the transition period (January 20−23), and the lockdown period (January 24−February 14, including an overlap from January 24 to 30 of the Spring Festival holiday). Multiple high-resolution traffic data were integrated during the research periods, including hourly traffic speeds within the sixth ring road for thousands of road links from AMap (Figure 1A and Figure S8), trajectory data from more than one hundred thousand floating cars (vehicles equipped with GPS) (Figure 1B), and estimated traffic flow by the SLOVE model (Figure S9a). Four days before the Spring Festival (transition period), people began returning to their hometowns, and the traffic condition became better with the average road speed increasing by 5−14%. During the COVID-19 lockdown period, the estimated traffic flow in freeways and urban roads dropped by 37−60% compared with the Pre period, with the traffic speed further increased by 14−31%, especially for roads within the fifth ring road during morning rush hours. Average daily vehicle kilometers of travel (VKT) of light duty vehicles (LDVs), heavy duty vehicles (HDVs), light duty trucks (LDTs), and HDTs decreased by 28, 61, 37, and 55%, respectively, during the lockdown period. After the 7 day Spring Festival holiday, the activity of LDTs gradually increased in order to meet the urban demand, but it was still 29% lower than the level during the Pre period. Compared with the data within the same period around the Spring Festival in 2019, the traffic speed affected by the pandemic remained at a higher level after the 7 day holiday during the lockdown period, with 6−14% higher than that in 2019 excluding snowy days (Figure S9b). These increases in traffic speed and decreases in traffic flow in Beijing, for such a long time, were significantly more marked than any previous holidays in the past few years.35

Based on the SLOVE model and TrackATruck model, the real-time on-road emissions of Beijing were calculated for the Pre period, the transition period, and the lockdown period. As a consequence of the COVID-19 pandemic, vehicle emissions decreased by 51−76%, with particularly a high reduction (76%) for NOx emissions (Figure 1C, other pollutants are shown in Figure S10). The relative reduction of emissions from the transportation sector was larger than the averaged decrease of all sectors, which was decreased by 21−69% during the lockdown period. The spatial distribution showed that in the lockdown period, vehicle emissions decreased substantially on almost all roads, especially for ring roads during the traffic rush hours and the main freight channels at night (Figure S11). The diurnal variations of vehicle emissions also showed a significant change during the lockdown period (Figure S12). Two emission peaks were observed on both weekdays and weekends during the Pre period, with the highest hourly NOx emissions reaching up to 9.2 Mg/h at 17:00 on weekdays. However, during the lockdown period, the hourly on-road emissions showed much smaller variations and the difference between weekdays and weekends became smaller.
NOx emissions at the evening traffic peak declined to 1.8 Mg/h. To sum up, our results indicate that the COVID-19 outbreak led to a significant reduction in traffic activities and of emissions compared with those in the Pre period and also changed the spatial distribution and diurnal variations of vehicle emissions.

In this study, the Pre period vehicle emission estimates were at the same magnitude with the recent Chinese government-led research (Figure S13). Before the COVID-19 lockdown, on-road emissions were estimated to be 496.8 Mg/day (CO), 99.6 Mg/day (HC), 165.1 Mg/day (NOx), and 5.1 Mg/day (PM2.5), accounting for 46, 29, 66, and 21% of the total anthropogenic emissions, respectively. HDTs were only responsible for 13 and 10% of NOx and PM2.5 emissions from all vehicles, much lower than previous estimations, because of the implementation of a HDT low emission zone in Beijing. Compared with estimations for 2013 based on a similar bottom-up method, NOx and PM2.5 emissions from on-road traffic reduced by 44–49% and 52–55%, indicating the effectiveness of the continuous vehicular emission control measures in recent years. All these features influenced the role of local traffic emissions in air pollution, which was comprehensively considered in this study.

Changes in Characteristics of Air Quality during the Lockdown. Figure 2A shows the observed temporal variations of daily AQI and PM2.5 concentrations before and during the COVID-19 outbreak, respectively. The haze pollution became more severe during the lockdown period compared with the one in either the Pre period or the transition period, with the mean daily PM2.5 level unexpectedly increasing from 48.0 to 99.0 μg/m³. Moreover, in the lockdown period, half of the days experienced high levels of pollution with daily PM2.5 concentrations exceeding 75 μg/m³, according to the level II standard of the Chinese National Ambient Air Quality Standards. The PM2.5 level remained at more than 150 μg/m³ for two episodes from January 25 to January 28 and from February 11 to February 13 (the first of these was excluded from this analysis because it was probably caused by fireworks). The variation in PM2.5 concentrations on polluted days was seen as an asymmetric “sawtooth” pattern, rising slowly before 2 days mainly driven by the gradual formation of secondary inorganic aerosols and then falling abruptly. In both the Pre period and lockdown period, these “sawtooth” periods including the stage of a gradual increase in concentrations were selected as heavy pollution periods (HPPs), and other consecutive clean days (daily PM2.5 level less than 75 μg/m³) were defined as nonheavy pollution periods (NHPPs). The unfavorable meteorological conditions during the HPPs were different from those in the NHPPs: low wind speed and planetary boundary layer height restraining the local air pollutant dispersion and high RH promoting the PM2.5 secondary formation (Figure S14). In this study, we separate episodes of HPPs and NHPPs to make comparisons between the Pre period and the lockdown period.

Figure 2A also shows the time series of the secondary aerosol enhancement, using the ratio of PM2.5 major secondary components (including sulfate, nitrate, ammonium, and SOM) to the EC, to eliminate the

![Figure 3. Changes in modeling concentrations between the real-time emission scenario (BASE) and the traffic-as-usual emission scenario (S1) during the lockdown period (BASE–S1). (A) O3, (B) NO3 radical, (C) SNAO (the sum of sulfate, nitrate, ammonium, and SOM), and (D) PM2.5. The black lines represent the ring roads of Beijing. Plots are for the model surface level.](https://dx.doi.org/10.1021/acs.est.0c04941)
impacts of different mixing conditions on the variation of observed pollutant concentrations.\textsuperscript{41} Here, we used the ratio of OM to EC to represent the secondary production of organic aerosols because the SOM dominated OM in Beijing on winter days with high RH and haze events\textsuperscript{42,43} and a field measurement around the COVID-19 pandemic has also proved the dominant role of SOM in the OM.\textsuperscript{44} The peak of the SNAO/EC ratio was not always lined up with the highest PM\textsubscript{2.5}, level during the polluted periods, especially for the HPP II, mainly due to the gradual loss of SNAO precursors and changes in the meteorological conditions (e.g., the direction of transported air masses). The ratios of SNAO to EC were stable between the Pre period and the transition period, while a significant rise was found during the lockdown period under either the HPP or the NHPP, with an average increase of 52% compared with the Pre period. We further investigated the changes in the diurnal variations of SNAO/EC between the Pre period and the lockdown period (Figure 2B). The enhancement of secondary aerosols during the COVID-19 outbreak was evident during the entire day with a peak level in the early morning (9:00–10:00 a.m.). This was especially true for nitrate aerosols, which was presented at a peak level more than twice than the one observed in the Pre period.

In spite of the increases in SNAO during the lockdown period, the concentrations of NO\textsubscript{3} and SO\textsubscript{2}\textsubscript{x} regarded as the two major gas-phase precursors of secondary PM\textsubscript{2.5} (nitrate and sulfate), declined by 30 and 35% on average, respectively (Figure 2C). Focusing on the differences between NHPPs, the relative reduction in NO\textsubscript{3} concentrations reached 58% during the lockdown period compared with the Pre period. This was consistent with the relative reduction of the estimated primary emissions (69%), indicating that the large emission reduction of the local anthropogenic sources actually decreased the PM\textsubscript{2.5} precursor concentrations during the COVID-19 outbreak.

Ozone is one of the most important oxidants in tropospheric chemistry. As shown in Figure 2C, the observed surface ozone increased by up to 263% between HPPs during the lockdown compared with the Pre period, with a period-averaged rise of 62%. In addition, we used a proxy O\textsubscript{3}−NO\textsubscript{2} to investigate changes in the nitrate radical (NO\textsubscript{3})\textsubscript{x}, the primary oxidant for nighttime secondary aerosol formation.\textsuperscript{45−47} The change in diurnal variations of O\textsubscript{3}, NO\textsubscript{3} radical, and NO\textsubscript{3} concentrations is provided in Figure S15. Compared with the Pre period, NO\textsubscript{3} radical concentrations also increased especially at night during the lockdown period. These changes during the COVID-19 outbreak and the subsequent lockdown indicate that the increased concentrations of oxidants facilitated the chemical formation of secondary fine particles in spite of the significantly reduced gaseous precursors, particularly resulting in a fast nitrate growth during the nighttime. In addition to the observations, our WRF-CMAQ modeling results (BASE scenario for the real-time simulation) also revealed a significant increase of the predicted oxidant concentrations in most areas of Beijing during the lockdown (Figure S16). Therefore, the significant enhancement of the local oxidizing capacity was partially responsible for the rapid growth of secondary aerosols during the lockdown period.

**Impacts of Vehicular Emission Reduction on Air Pollution during the Lockdown.** Figure 3 shows the changes in spatial distribution of the predicted oxidants and PM\textsubscript{2.5} concentrations between BASE and S1 from WRF-CMAQ modeling results, reflecting the impacts from reduced vehicle emissions on air quality during the lockdown. In the analysis of modeling results, the predicted SOM, rather than the total OM, was included in the SNAO concentrations to directly represent the secondary formation of OM. A significant increase in the O\textsubscript{3} concentration of up to 11 parts per billion by volume (ppbv) was seen in urban areas within the sixth ring road and southern areas, induced by the larger vehicular NO\textsubscript{x} emission reduction compared with VOCs under the VOC-limited ozone formation regime.\textsuperscript{48,49} Compared with Figure S16a (NHPP II – NHPP I), which shows the increased O\textsubscript{3} from the Pre to the lockdown period between NHPPs when the concentrations were more affected by local sources, the O\textsubscript{3} enhancements only caused by vehicle emissions are similar (Figure 3A). It could be inferred that local traffic emission reduction is the main driving force for the enhanced oxidation capacity in urban areas of Beijing. Compared with changes in O\textsubscript{3} concentrations, the increase in NO\textsubscript{3} radical concentrations was relatively small at the surface level because of the sharp decline of ambient NO\textsubscript{2} concentrations (Figure 3B), while a more obvious increase was found in the upper air within urban areas (approximately 46 m above the ground) due to weaker NO titration effects (Figure S17). The enhanced oxidants facilitated the formation of SOM during the day and nitrate aerosols in nighttime (Figure S18). However, the increased secondary aerosol concentrations were small, with a slight rise by up to 1.6 μg/m\textsuperscript{3} (Figure 3C), probably because of the inadequate precursors particularly during the lockdown period. Meanwhile, the increased secondary aerosols were still enough to offset the benefit of vehicular reductions in primary emissions, leading to a modest increase in the total PM\textsubscript{2.5} concentrations by up to 1.4 μg/m\textsuperscript{3} (Figure 3D).

Our simulations show that the spatial variations of the atmospheric oxidation induced the opposite changes in PM\textsubscript{2.5} formation in rural (outside the sixth ring road, except for the south) compared with urban areas. The rise of O\textsubscript{3} in rural areas was relatively small because 81–83% of the vehicle emission reductions in Beijing occurred in urban areas and the ozone formation regime changed from VOC-limited to NO\textsubscript{x}-limited going from urban to rural areas.\textsuperscript{48,49} In addition, the reduction of gas-phase precursors restricted the production of secondary particles in rural areas, resulting in the decrease of SNAO and PM\textsubscript{2.5} concentrations. Such contrasting impacts between urban and rural areas on both SNAO and PM\textsubscript{2.5} were more distinct during the HPP compared with those during the NHPP, mainly due to unfavorable meteorological conditions for air pollutant dispersion during the HPP.\textsuperscript{40} Moreover, the region with the evident increase of oxidants (urban areas) was different from that with the evident increase of SNAO or PM\textsubscript{2.5} (south rural areas) due to the influence of regional transport. An enhanced flux of PM\textsubscript{2.5} precursors from the Hebei province, a populated and industrial region, brought by prevailing southerly wind during the pollution episodes, particularly increased the secondary aerosol formation caused by the increase of oxidants in downwind south rural areas of Beijing.\textsuperscript{50}

As a conclusion here, the imbalance in emission reduction of NO\textsubscript{x} and VOCs from vehicles was an important cause for the rise of local atmospheric oxidizing capacity, resulting in a modest increase of secondary aerosols and PM\textsubscript{2.5} concentrations. However, the slightly increased PM\textsubscript{2.5} induced by the considerable vehicular emission reduction could not explain the observed significant growth of secondary aerosols in Beijing during the COVID-19 lockdown period.
The contribution of regional transport to PM$_{2.5}$ concentrations in Beijing. The percentages in the top of the figure indicate the contribution of regional transport to PM$_{2.5}$ concentrations in Beijing during NHPPs and HPPs.

Figure 4. Simplified schematic of the role of reduced vehicle emissions and regional transport in the "pandemic haze" formation in Beijing. The percentages in the top of the figure indicate the contribution of regional transport to PM$_{2.5}$ concentrations in Beijing during NHPPs and HPPs.

The general public was disappointed by the "pandemic haze" because it failed the expectation that traffic control measures would prevent PM$_{2.5}$ pollution from happening in Beijing. Our modeling results indicate that, however, the winter haze events in Beijing were insensitive to the local traffic emission reductions. Based on a series of sensitivity analysis modifying the local on-road emissions on winter normal days (scenarios S2–S6), we investigated the response of PM$_{2.5}$ and oxidants to vehicle emission changes, while other nonvehicle sectors remained at their usual level. The results showed that even without vehicle emissions, the ambient predicted PM$_{2.5}$ concentrations could only be reduced by 1.2 µg/m$^3$ on average in urban and southern rural areas of Beijing (Table S5). Although vehicles accounted for 66% of the local NO$_x$ emissions, the predicted concentrations of oxidants and fine particles would experience a consistent increase, with the reduction ratio of the vehicular NO$_x$ emissions rising from 0 to 100%, even with a 100% reduction in VOC emissions from vehicles (Table S5). On the one hand, the abundant nitrate precursors in neighboring regions suppress the effectiveness of local NO$_x$ emission control. On the other hand, reducing local NO$_x$ emission favors the enhanced atmospheric oxidation ability to form more secondary particles because many urban areas in China are prevailing under the VOC-limited condition. Although reducing vehicular VOC and primary PM emissions were both positive in decreasing the PM$_{2.5}$...
levels, unfortunately, traffic control usually leads to a greater NO\textsubscript{x} reduction than VOCs, which goes to explain why the annual reduction of vehicular emissions had resulted in reductions in VOCs that represent only half (in percentage terms) the NO\textsubscript{x} reductions over the past years.\textsuperscript{55} All these points above explain why traffic control cannot mitigate the winter haze pollution in Beijing at the present time, and this question also needs to be better explained to the general public.

In the past years, the gradual strengthened vehicle emission controls have successfully contributed to the PM\textsubscript{2.5} decrease in China.\textsuperscript{51} In addition, past experiences from developed countries indicate that emission control on the continuously growing motor vehicle fleet is efficient and ultimately cost-effective to relieve the air pollution in a megacity.\textsuperscript{9,10} The current problem is the imbalanced control among different source regions and kinds of air pollutants. Compared with its surrounding regions, local source emissions in Beijing were reduced faster in the past years, which led to the share of regional transport for the air pollution in Beijing increasing to 76\% on average during winter (Figure S20). As for the differences in local species controls in Beijing, the ratio of annual averaged emission reduction of VOCs to NO\textsubscript{x} used to be approximately 1.2 (23.4 kt vs 28.9 kt) from 2013 to 2017.\textsuperscript{7} However, this VOCs/NO\textsubscript{x} ratio is still small enough to increase the atmospheric oxidizing capacity under the strong VOC-limited condition in winter,\textsuperscript{62} which is proved by the enhanced oxidants during the COVID-19 lockdown (total daily emission reduction of VOCS/NO\textsubscript{x} is also 1.2, 217.7 Mg vs 174.3 Mg, see Figure 1C). Therefore, the key is not judging whether traffic emission control is necessary but accelerating VOC emission reductions and synchronously controlling regional sources to release the benefits of local traffic emission control.

Targeting any of long-term air quality, climate change, or street-level personal exposure, any measures reducing vehicle emissions are going to be beneficial. To achieve PM\textsubscript{2.5} concentration reductions in the short term, VOCs and primary PM\textsubscript{2.5} should be jointly treated as the prior pollutants to control, or NO\textsubscript{x} emissions should be substantially reduced by the combination with other sources beyond vehicles so as to reach the nonlinear tipping point between changes in NO\textsubscript{x} reductions and oxidant concentrations. For the first option, the new emission standard for LDVs (China 6), which was implemented in July 2020, is expected to dramatically reduce the VOC emissions from evaporation and will be effective to improve the air quality.\textsuperscript{63} The challenge is to maintain popular support for mitigation policies such as reductions in traffic flow or restrictions in vehicle type, which themselves lead to significant air quality improvements (PM\textsubscript{2.5}, NO\textsubscript{x}) but which are not directly visible to the general population in the face of the highly visible haze events.

Our study was subject to a few uncertainties and limitations. In addition to uncertainties of emission inventories and WRF-CMAQ models (discussed in the Supporting Information sections S4 and S5), the SOM was not considered in the source apportionment due to the limitation of the existing ISAM. We probably underestimated the contribution of local vehicle emissions to PM\textsubscript{2.5} concentrations because (a) aromatics from gasoline vehicles and IVOCs from both gasoline and diesel vehicles are critical determinants of urban secondary organic aerosol formation\textsuperscript{64−66} and (b) synergic oxidation of vehicular exhaust leads to efficient formation of ultrafine particles under urban conditions.\textsuperscript{67} Moreover, the on-road emissions are instantly diluted throughout the coarse grid cells of the chemical transport model in which the emissions occur. In a finer spatial scale, however, the vehicle emission would significantly affect the human exposure to air pollution due to its close proximity to human activities at a low emission height and thus result in a serious health burden.\textsuperscript{58} Quantification of vehicular contribution to human health risk in Beijing at a neighborhood scale based on a source−receptor model with a higher spatial resolution is necessary and suggested for future investigation.
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