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Substantial Changes in Nitrogen Dioxide and Ozone after Excluding Meteorological Impacts during the COVID-19 Outbreak in Mainland China

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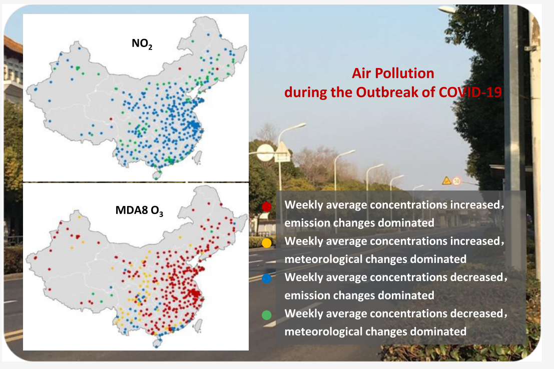
ABSTRACT: The COVID-19 outbreak in China led to dramatic changes in human activities resulting from the sudden infection prevention and control measures. Here, we use ground-level observations and model simulations to examine the nationwide spatial−temporal variations of six air pollutants before and after the initiation of First-Level Public Health Emergency Response. The level of ambient NO_2 declined significantly, and in most cities, the decline was dominated by reduced emissions. Meanwhile, the level of O_3 increased significantly during this period, and the nonmeteorological factors explained the increase. For the other air pollutants ($PM_{2.5}$, SO_2, and CO), the observed declines on the national scale were obviously affected by the meteorological conditions. In Wuhan, significant declines were found for air pollutants except O_3 and emissions dominated the changes, while in Beijing during the same period, only the level of NO_2 significantly declined. This study clearly shows that the meteorological changes contributed substantially to the observed changes in most air pollutants, and this must be considered in evaluating the impacts of pollutant source changes on air quality during the specific event and in assessing source-oriented risks.



■ INTRODUCTION

Air pollution is one of the most concerning environmental challenges in China, which causes more than 1 million premature deaths each year.[1−3] Although considerable effort has been devoted to mitigating air pollution during the past several decades, many cities still suffer from severe air pollutions. For instance, Beijing's average $PM_{2.5}$ concentration was 42 $\mu g/m^3$ in 2019.[4] It is close to China's official air quality standard (35 $\mu g/m^3$) but still far short of the World Health Organization (WHO)'s guideline value of 10 $\mu g/m^3$.[5] In fact, there were 180 of 337 cities (53.4%) monitored in mainland China exceeding China's official $PM_{2.5}$ standard in 2019.[4]

The outbreak of 2019 novel coronavirus (2019-nCoV) in mainland China occurred reportedly in December 2019.[6,7] The rapid growth in the confirmed case number and deaths led to a series of preventive anti-epidemic measures implemented by the Chinese government. From January 23 to 29, 2020, China activated the First-Level Public Health Emergency Response (FLPHER), which implemented strict travel restrictions involving more than 1.3 billion people.[8] As a consequence, transportation, construction, and light industry activities were dramatically decreased. For example, the total traffic flow across China decreased by 87.7% on January 30, 2020, compared with the same period last year.[9]

Such an event had significant impacts on the social−economic activities and thus on the environment.[10] It is therefore interesting, using the national air quality monitoring network,[11] to look into the changes in nationwide air quality over such a short period. In addition, evaluating the changes in air pollutant concentrations with the exclusion of meteorological impacts is crucial. This study is expected, as a preliminary evaluation, to provide important information about the air quality changes affected by FLPHER due to the COVID-19 outbreak.

■ MATERIALS AND METHODS

Study Period and Data Recording. The FLPHER activation dates were from January 23 to 25 for 30 provincial-level regions in China. Therefore, to look into its impacts on short-term air quality, two time periods, one week before (January 16−22, 2020, period I) and one week after

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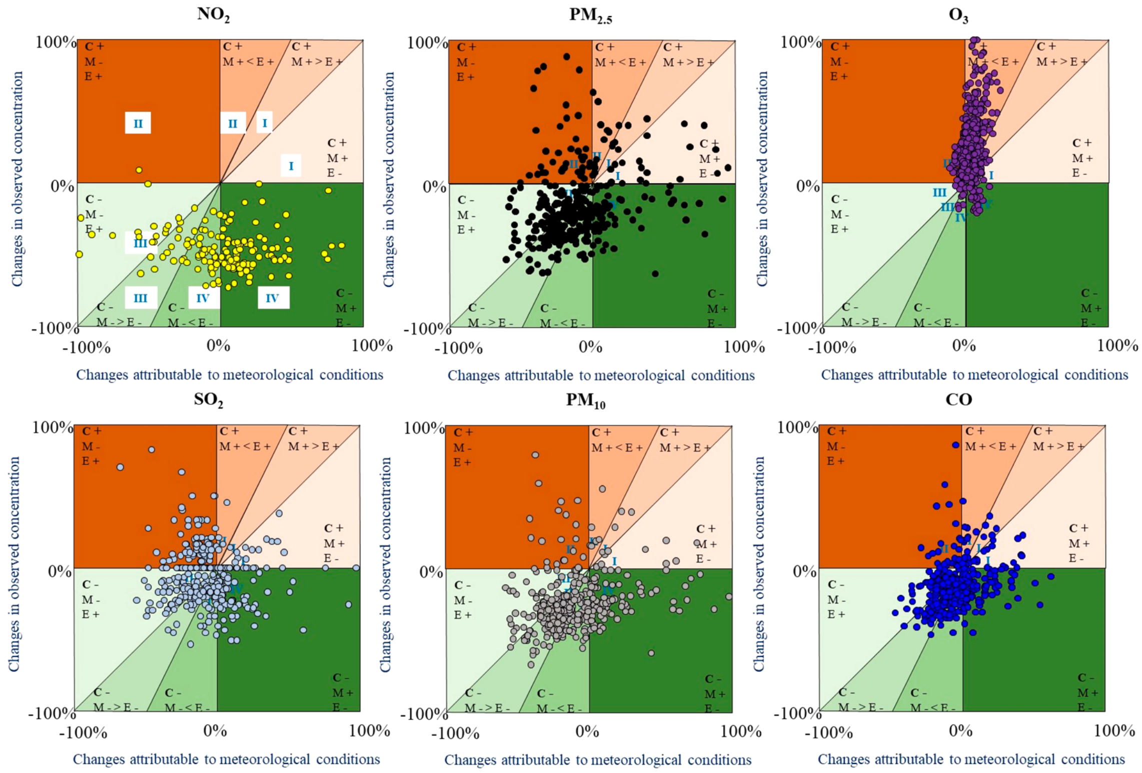

Figure 1. Changes in the observed weekly average concentrations between the two periods vs the changes that can be attributed to the meteorological differences. In the first and second quadrants, ambient concentrations increased regardless of the meteorological changes, and in the third and fourth quadrants, the weekly average ambient concentrations declined. In region I, the meteorological changes dominated the ambient concentration increases. In region II, emission changes dominated the ambient concentration increases. In region III, the meteorological changes dominated the ambient concentration decreases. In region IV, emission changes dominated the ambient concentration decreases. C+ and C−: observed weekly average concentrations increased and decreased, respectively. M+ and M−: meteorological differences between the two periods resulted in increases and decrease, respectively, in air pollutant concentration based on the model simulation (note that the model has difficulty in accurately evaluating the boundary layer resulting in the uncertainties given here). E+ and E−: emission changes were indicated to increase and decrease, respectively, the weekly average concentrations of air pollutants (the differences between observed and simulated weekly average concentration were positive and negative, respectively).

(January 26 to February 1, 2020, period II) the initiation of FLPHER, were covered for these 30 provinces consisting of 354 cities. Tibet activated FLPHER on January 29, 2020, which was several days later than other 30 provinces.[12] Therefore, the time periods for Tibet were postponed accordingly (January 22−28 as period I and January 30 to February 5 as period II).

The monitoring data of six air pollutants (i.e., $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2, and O_3) were obtained from the China National Environmental Monitoring Center (CNEMC) (http://106.37.208.233:20035/). Detailed information about the monitoring system was described previously.[11] In total, 361 cities with 1632 monitoring sites from 31 provinces that activated FLPHER were included in this study. The details of research regions and the activation dates are listed in Table S1. For each period, the average concentrations of $PM_{2.5}$, PM_{10}, CO, SO_2, and NO_2 were calculated as the means of daily 24 h average concentrations, and the average concentration of O_3 was calculated as the mean of the maximum daily 8 h average concentrations.

Data Analysis and Statistics. Data statistical analysis was performed using SPSS version 20.0 (SPSS, Chicago, IL). Results are given as the mean ± the standard deviation (SD). Differences were considered significant if $p < 0.05$, unless otherwise specified.

Air Quality Modeling. It is well documented that meteorological conditions play an important role in ambient air pollution via multiple pathways involving emission, transport, chemistry, and deposition.[13,14] In this study, the Weather Research and Forecasting (WRF) model and the Community Multiscale Air Quality (CMAQ) model[15−20] were used to evaluate the impacts of the changing meteorology on air quality before and after FLPHER was activated. WRF was driven by the fifth generation of the European Centre for Medium-Range Weather Forecasts atmospheric reanalysis (ERA5)[21] and provided meteorological inputs for the CMAQ simulation. The WRF model performance was evaluated, showing good agreement with ground-level observations (Figure S1). The CMAQ simulation covered the entire mainland China at the 36 km horizontal resolution and spanned the period from January 8, 2020, to February 6, 2020, covering all days in periods I and II with the first 8 days for spinning up. To evaluate the changes in air pollution associated with the meteorological conditions, the real-world meteorology informed by the ERA5 reanalysis (Table S2) was used as inputs in the model simulation with fixed emissions for 2017 because the real-time emissions for the study period were not available. This brings in considerable uncertainties in model results as pollutant emissions in China are changing,[20] and moreover, significant changes were expected during the specific COVID-19 period. The model here is primarily run to

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isolate impacts of meteorology and emissions, and the results of the relative changes in air pollutant concentrations from the modeling were adopted in subsequent analysis. Thus, the day-to-day variations for the simulated concentrations of air pollutants were attributed to the impacts of day-to-day meteorological changes as daily emissions were held constant over the simulation period. The variations for observed pollutants concentrations were attributed to the mixed impacts of meteorological changes and emission changes. Subsequently, upon exclusion of the meteorological impacts modeled by CMAQ from the observed overall changes in pollutant concentrations, the emission impacts on air quality between the two periods were isolated. The boundary layer plays an important role in air pollution formation and dispersion; however, it should be noted that WRF and other regional models have difficulties in simulating the evolution of the boundary layer.[22,23] Therefore, the impacts of meteorological factors may not be completely and accurately captured by the model. The model configurations and performance are detailed in the Supporting Information.

RESULTS AND DISCUSSION

Nationwide Air Pollutant Concentrations. The nationwide average concentrations of air pollutants during the two time periods, before and after the initiation of FLPHER, were significantly different ($p < 0.01$). In period I, the average concentrations of $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2, and O_3 were 67 ± 39, 88 ± 47, 1120 ± 390, 13 ± 10, 32 ± 14, and 65 ± 14 $\mu g/m^3$, respectively. After the initiation of FLPHER (period II), they were 56 ± 34, 67 ± 39, 970 ± 380, 12 ± 9, 17 ± 9, and 82 ± 11 $\mu g/m^3$, respectively. The concentrations decreased by 13.7%, 21.8%, 12.2%, 4.6%, and 46.1% on average for $PM_{2.5}$, PM_{10}, CO, SO_2, and NO_2, respectively, whereas an increase in 47.3% was observed in O_3. As expected,[24−26] $PM_{2.5}$ correlated positively with PM_{10} (r = 0.904; $p < 0.001$). The ratios of $PM_{2.5}$ in PM_{10} were >0.60 in most cities, indicating the high impact of secondary formation.[26,27] A significant negative correlation was observed between the changes in the concentration of NO_2 and O_3 (r = −0.45; $p < 0.001$) (Figure S4), indicating important roles of VOCs in ozone formation in these cities during the study period. Ozone formation is complicated by its association with the abundance of NOx and VOCs.[28] At this stage due to the absence of the national VOC concentrations, it is hard to further explore the relationship and formation mechanism(s) here, but it is imperative to look into the process in future work, to evaluate the effect of human activities on ozone pollution and adverse health impacts of ozone exposure. Previous studies also indicate that the decrease in the $PM_{2.5}$ concentration may also play a role in the increase in the level of ozone;[29] however, there was no significant correlation found between O_3 and $PM_{2.5}$ in this study (r = −0.03), which may be related to the differences between winter and summer.

Meteorological Impacts on Air Pollutants. The WRF simulation showed a large-scale increase in temperature over northern China, especially in Inner Mongolia, during period II compared to period I (Figure S5), which could potentially lead to enhanced photochemical formation of ozone and secondary PM,[30,31] and showed decreases in wind speed and boundary layer height in this region (Figure S5), which could favor the accumulation of pollutants.[30] On the other hand, a decrease in temperature and increases in wind speed and boundary layer height were found in southern China (Figure S5), indicating inhabited ozone formation and a favorable ventilation condition. The change in precipitation was minor in most parts of China (Figure S5) given the ongoing dry season.

Figure 1 plots the observed concentration changes against meteorology-induced changes for the 361 cities. As one can see, the observed NO_2 concentrations declined in nearly all cities, while in 53% of these cities, the meteorological conditions would even increase the NO_2 levels if emissions were not changed (the fourth quadrant in Figure 1). Only in 20% cities were the NO_2 declines primarily a result of the meteorological changes (regions III in Figure 1). Our analysis suggests that changes in emissions contributed to the observed NO_2 declines in 91% cities (the area below the 1:1 line) and were the dominant cause of the NO_2 declines in 80% of the cities (region IV). For O_3, obvious increases were observed in most cities. In only 6% of the cities, the increases were primarily attributable to the meteorological changes (region I in the first quadrant), indicating that the increases in the level of O_3 in most cities were due to the changes in precursor emissions. In approximately 20% of the cities, although changes in the meteorological conditions would result in decreases in O_3, the changes in precursor emissions and also probably the nonlinear response of ozone to the precursors during the atmospheric reactions were found to overwhelm the changes in meteorology, ultimately resulting in increases in the level of O_3 (the second quadrant). The 10 cities showing the largest increases in the level of O_3 are all located in the North China Plain, one of the most populous regions in China, indicating a potential negative health effect from O_3 exposure in this region. Note that the changes in O_3 in these cities were all dominated by emission changes. On the other hand, some cities showed decreases in the level of O_3 due to emission changes, and these cities are mainly located in southeastern China (shown in Figure S6).

For $PM_{2.5}$, among the 287 cities (of 361, 80%) with decreased levels of $PM_{2.5}$, 60% of the cities were mainly due to the meteorological impacts. There were 22% cities where the meteorological differences would increase the $PM_{2.5}$ levels between the two periods, but changes in emissions ultimately resulted in the observed declines. The situation was generally very similar for SO_2, CO, and PM_{10}; 20%, 15%, and 10% of the cities had higher weekly average SO_2, CO, and PM_{10} concentrations, respectively, shortly after the restriction, and in those with decreased concentrations observed, the meteorological changes overwhelmed the emission impacts leading to concentration declines of 57%, 44%, and 53% of the cities, respectively. As shown in Figure S6, for $PM_{2.5}$ and PM_{10}, cities with similar changes in observed weekly concentrations were geographically close. Cities with increased levels of $PM_{2.5}$ and PM_{10} were mainly located in the northeastern (Liaoning and Jilin provinces) and southwestern (Yunan, Guizhou, and Guangxi provinces) China, and the changes were mainly attributed to nonmeteorological impacts.

As mentioned above, the real-time emission estimates for the study period were not available when we worked on this study, and the fixed emissions from 2017 as model inputs may bring in considerable uncertainties in the results, although only the relative changes (percentage) in the modeled air pollutant concentrations were used. We, as a proxy to test the robustness of results, further run a sensitivity test by using a different model (the Weather Research and Forecasting model coupled with Chemistry, WRF-Chem)[15] with a different emission inventory (2014 instead of 2017) to evaluate the impacts of

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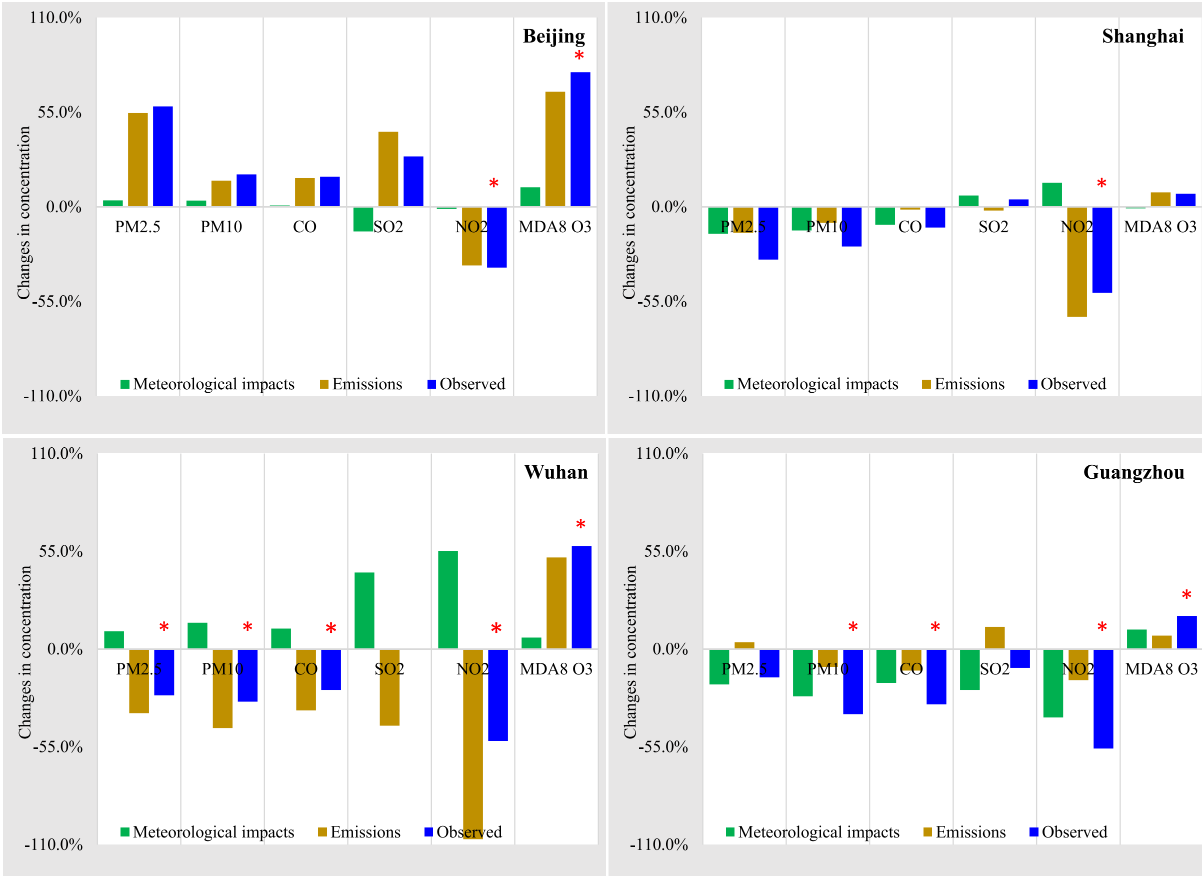

Figure 2. Changes in the observed weekly average concentrations between the two periods against the changes attributable to the meteorological and emission changes for the four representative cities.

real-time meteorology on air pollution during this period. This modeling platform had been previously used in evaluating the impacts of residential emissions on air pollution in China, and previous works did reveal acceptable results from the modeling platform in comparison with the observation.[32,33] The WRF-Chem simulated relative changes in air pollution were generally similar to that from the WRF-CMAQ modeling, as shown in Figure S7 and Figure 2. Therefore, it could be inferred that the observed changes in the short-term air concentrations of $PM_{2.5}$, PM_{10}, CO, and SO_2 in nearly half of monitored cities in the mainland China, and also NO_2 in some cities (20%) during the study period, were primarily due to the meteorological changes, while for the increased O_3 levels, emissions played a more important role. In fact, compared to January 2019, although the monthly electricity consumption of January 2020 for production and supply of electricity and heat decreased by 30%, there were only slight decreases observed for petroleum, coal, and other fuel processing industries (5.5%), the ferrous metal smelting and rolling processing industry (5.6%), and the nonferrous metal smelting and rolling processing industry (2.3%) (https://www.wind.com.cn/). The weekly operating rate of Chinese blast furnaces decreased by only 4.5% during FLPHER, compared to the week before the activation of FLPHER, and the weekly refinery utilization rates decreased by only 3.5% (https://www.wind.com.cn/). These indicated that primary air pollutant emissions associated with these activities did not decline much.

Air Pollutants for Four Representative Cities. Here we specifically examined the temporal changes in air pollution in four representative cities, including Beijing, Shanghai, Guangzhou, and Wuhan (Table S3). These four megacities are located in northern, central, eastern, and southern China, respectively. Beijing, Shanghai, and Guangzhou are three megacities in the Beijing−Tianjin−Hebei, Yangtze River Delta, and Pearl River Delta three-megacity clusters, respectively, which are target areas in the plans of the Chinese government to mitigate air pollution specifically (www.mee.gov.cn/hjzl/dqhj/cskqzlzkyb/). Different temporal variations and distinct meteorological and emission impacts could be found among four cities. In Beijing, the weekly average concentrations of most air pollutants except NO_2 were higher during period II than during period I, and the changes can be primarily explained by emissions. During the 2008 Olympic Games held from August 8 to 24 and the APEC summit of 2014 held from November 7 to 12, the concentrations of all six air pollutants decreased in Beijing due to stringent controls. During those two events, except for the transportation restrictions, some polluting factories in surrounding regions were also closed. During FLPHER, on the other hand, there was no intentional restriction on heavy industry.[34] Additionally, different from those two previous events, the outbreak of COVID-19 occurred during cold seasons when space heating was needed in northern China. It has been well documented that fuel burning for space heating, especially in low-efficiency in-home heating stoves that burn coal and wood in the surrounding rural areas, significantly affects air quality.[35]

In the other three cities, the weekly average concentrations of $PM_{2.5}$, PM_{10}, and CO were lower during period II than

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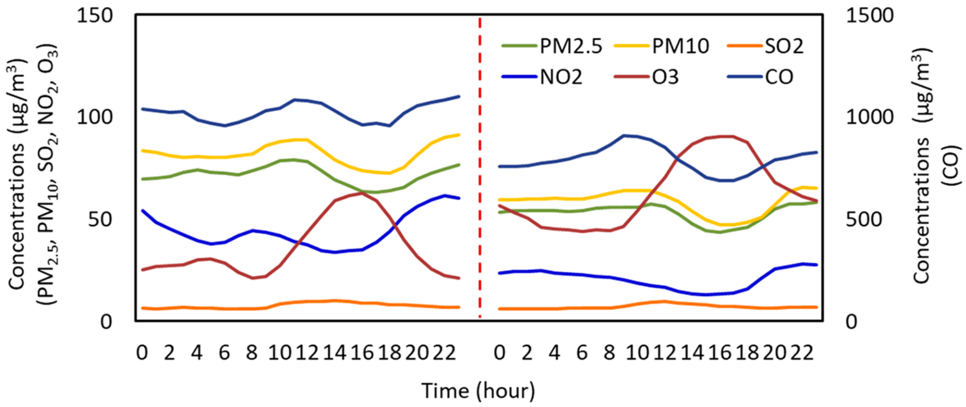

Figure 3. Diurnal variations of $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2, and O_3 before and after the initiation of the FLPHER, illustrated by the data collected from Wuhan. The left and right sections show the diurnal variations before and after the initiation of FLPHER, respectively.

during period I. In Shanghai, located in eastern China, the level of NO_2 significantly decreased, by 63.9%, primarily due to changes in emission. In Guangzhou, another megacity located in southern China, the levels of PM_{10}, CO, and NO_2 significantly decrease by 44.8%, 36.8%, and 77.7%, respectively, and meteorological impacts dominated the declines. Different from the other three cities, in Wuhan, where the earliest preventive measures were conducted due to the outbreak of COVID-19, the levels of not only NO_2 but also $PM_{2.5}$, PM_{10}, and CO significantly declined by 31.6%, 34.7%, and 26.0%, respectively. Even though the meteorological changes were unfavorable, the decline in emission dominated the changes in this city, driving the decreases in the concentrations of all of the pollutants except O_3.

The controls affected not only the daily average concentrations but also the diurnal profiles. The diurnal variation in NO_2 was clearly different between the two study periods. With Wuhan as an example (Figure 3), the NO_2 concentration plateaued at a lower level and almost even sacrificed the daily variation mode after the initiation of FLPHER, resulting in the dramatic decrease in the level of NO_2. In addition to the presence of precursors, the O_3 level is strongly affected by ambient conditions like temperature and solar radiation; namely, the high temperature and radiation are favorable for O_3 formation.[36] After the initiation of FLPHER, O_3 concentrations still reached the peak at 17:00 as before, but at much higher levels. For $PM_{2.5}$ and SO_2, the diurnal profiles were similar before and after FLPHER.

In conclusion, this study investigated the nationwide air pollution characteristics during the outbreak of COVID-19 in mainland China. As a significant worldwide public event, the FLPHER action affected more than a billion people. We observed significant decreases in the concentrations for NO_2 in >85% of cities, and most of the declines can be attributable to changes in emission. This is consistent with the remarkable decrease in the level of urban traffic, a major source of NO_2, during FLPHER.[9] For O_3, precursor emissions were mainly responsible for the observed increase, while meteorological changes play a relatively minor role. Meanwhile, although the weekly average concentrations decreased in >75% of the cities for $PM_{2.5}$, PM_{10}, CO, and SO_2, most of the declines were not significant, indicating that except for NO_2, only by vehicle restrictions, such as the license plate restriction,[37] might not be able to significantly improve air quality in most study cities. The insignificant changes in other pollutants call for further evaluation of the source contributions to ambient air pollution.

It should be noted that the initiation of FLPHER happened to be at the Spring Festival of China, which also likely contributed to the large-scale changes in air pollution. In this study, we did not clearly differentiate the contributions of each source, which warrants further investigation. In addition, this analysis was conducted on the basis of the monitoring data from CNEMC and air quality modeling with fixed emissions. Follow-up studies should further quantify the influences of preventive measures on pollutant emissions by combining bottom-up and top-down emission information to assess the unintended impact of this event on air quality. The social event would be also an opportunity to study the atmospheric chemistry process in the formation of ozone in terms of changes in NO_2 and VOCs when the sources and observation concentrations of VOCs were available.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.0c00304.

Details of provincial-level regions affected by the FLPHER (Table S1), ERA5 meteorological variables used as initial and boundary conditions, nudging fields, or both to drive the WRF simulation (Table S2), evaluation of the WRF model performance (Figure S1), evaluation of the CMAQ simulation (Figure S2), comparison of daily variations in modeled and observed pollutant concentrations (Figure S3), correlations between air pollutant concentration changes at a nationwide scale (Figure S4), WRF-modeled changes between periods I and II (Figure S5), spatial distribution of cities with similar changes in concentration and changes attributed to meteorological or nonmeteorological impacts (Figure S6), correlation changes in the observed weekly average concentrations against the changes attributable to the meteorological differences modeled using the WRF-Chem model (Figure S7), and concentration changes in the air pollutants for four representative cities (Table S3) (PDF)

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Notes

The authors declare no competing financial interest.

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