Highly Resolved Dynamic Emissions of Air Pollutants and Greenhouse Gas CO₂ during COVID-19 Pandemic in East China

Cheng Huang,* Jingyu An, Hongli Wang,* Qizhen Liu, Junjie Tian, Qian Wang, Qingyao Hu, Rusha Yan, Yin Shen, Yusen Duan, Qingyan Fu, Jiandong Shen, Hui Ye, Ming Wang, Chong Wei, Yafang Cheng, and Hang Su*

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INTRODUCTION

The coronavirus (COVID-19) pandemic has triggered an unprecedented shock on global energy, industry, and transportation sectors.¹ China was affected by the virus and initiated lockdown since January 23, 2020.² The unintentional emission reductions caused by COVID-19 lockdowns provide an opportunity to quantify their synergetic effects on air pollutants and CO₂ emissions and their synergy. Here, we constructed an approach to estimate city-level high resolution dynamic emissions of both anthropogenic air pollutants and CO₂ by introducing dynamic temporal allocation coefficients based on real-time multisource activity data. We first apply this approach to estimate the spatiotemporal evolution of sectoral emissions in eastern China, focusing on the period around the COVID-19 lockdown. Comparisons with observational data show that our approach can well capture the spatiotemporal changes of both short-lived precursors (NOx and NMVOCs) and CO₂ emissions. Our results show that air pollutants (SO₂, NOx, and NMVOCs) were reduced by up to 31%−53% during the lockdown period accompanied by simultaneous changes of 40% CO₂ emissions. The declines in power and heavy industry sectors dominated regional SO₂ and CO₂ reductions. NOx reductions were mainly attributed to mobile sources, while NMVOCs emission reductions were mainly from light industry sectors. Our findings suggest that differentiated emission control strategies should be implemented for different source categories to achieve coordinated reduction goals.

NOx emissions³⁴⁻⁵ but did not include other pollutants [e.g., SO₂, nonmethane volatile organic compounds (NMVOCs), and primary PM₁.₅]. A recent study provides real-time estimations of emission changes using an emission concentration response model.¹⁷ However, its application is limited to estimate total emissions. Detailed source-specific estimates for individual industrial, transportation, and residential sectors are still lacking.

Another issue of concern is that there were currently few studies to systematically assess the simultaneous changes in greenhouse gases in the course of air pollution control. China is facing a huge challenge to reduce CO₂ emissions.¹⁸ It is critical to find an effective way to co-control greenhouse gases while reducing PM_{2.₅} and ozone pollution. Recent studies have developed high temporal resolution emission inventories using satellite retrieval¹⁹ and bottom-up approaches,²⁰,²¹ capturing the daily CO₂ emissions during the COVID-19 pandemic.

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Nevertheless, a high temporal resolution dynamic emission inventory which can be refined to city-level and specific sectors and include both CO₂ and air pollutants has not yet been reported.

Figure 1. Temporal variations of air pollutants and CO₂ emissions in the first quarter of 2019 and 2020 in the YRD region. SO₂ (a), CO (b), NOₓ (c), NMVOCs (d), BC (e), POC (f), and CO₂ (g). The black solid and thin dashed lines represent the daily emissions in 2019 and 2020, respectively. The shaded areas represent contributions of individual sectors to the differences of emissions between 2019 and 2020 (two years). The periods of Pre, LD, RS1, and RS2 are marked and separated by dotted lines.
Here, we constructed an approach driven by dynamic temporal allocation coefficients, which is based on real-time activity data from multisources. This approach can generate detailed anthropogenic sources in high temporal and spatial resolution, which was validated and applied to the Yangtze River Delta (YRD) region, one of the most densely populated area with densely distributed industries and transportation in China. The emissions were grouped into five major source categories, including power plants (Power), heavy industry (H-Ind), light industry (L-Ind), mobile sources (Mobile) and the others (Others), and 25 subcategories. Detailed classification is presented in Table S1. The pollutants include SO2, NOx, CO, NMVOCs, and primary particles such as PM2.5, black carbon (BC), and primary organic carbon (POC), as well as CO2. Changes of observed concentrations of air pollutants, like NOx and NMVOCs, as well as CO2 were used to validate the high-temporal resolution approach. Then, we evaluated the impacts of different source controls on air pollutants and CO2 emissions based on the validated inventory, providing an exemplary case for coordinated control of air pollutants and CO2 in China.

## MATERIALS AND METHODS

**Framework of Dynamic Emission Estimation.** Figure S1 shows the methodological framework. We first normalized the real-time activity data to establish a set of dynamic temporal allocation coefficients, then multiplied them by baseline emissions for each subcategory. The baseline emission inventory was developed in our previous study22 (see the Supporting Information). For energy-related industries, including Power and most sectors in H-Ind such as petroleum refining, coking, iron and steel, cement, and nonferrous metal manufacturing, we used hourly continuous emission monitoring system (CEMS) data and merged them into daily. Considering the CEMS data only monitored SO2, NOx, and PM emissions, we used SO2 as a proxy to substitute CO and CO2 emissions since they all depended mainly on fuel consumption, while PM, BC, and OC were all particulate matter, therefore substituted by PM. For industries involving NMVOC emissions, such as chemical manufacturing, textile, and industrial coating, and more, industrial electricity consumption data and monthly industrial production data to temporal allocation coefficients, which is based on real-time activity data from multisources. This approach can generate detailed anthropogenic sources in high temporal and spatial resolution, which was validated and applied to the Yangtze River Delta (YRD) region, one of the most densely populated area with densely distributed industries and transportation in China. The emissions were grouped into five major source categories, including power plants (Power), heavy industry (H-Ind), light industry (L-Ind), mobile sources (Mobile) and the others (Others), and 25 subcategories. Detailed classification is presented in Table S1. The pollutants include SO2, NOx, CO, NMVOCs, and primary particles such as PM2.5, black carbon (BC), and primary organic carbon (POC), as well as CO2. Changes of observed concentrations of air pollutants, like NOx and NMVOCs, as well as CO2 were used to validate the high-temporal resolution approach. Then, we evaluated the impacts of different source controls on air pollutants and CO2 emissions based on the validated inventory, providing an exemplary case for coordinated control of air pollutants and CO2 in China.

**Ground-Based Observation.** NO2 concentrations were obtained from the air monitoring data center of Ministry of Ecology and Environment of China. The data covered 180 monitoring sites in 41 cities of the YRD region. Their locations are shown in Figure S2. The NMVOC species were all observed by custom-built online gas chromatography systems equipped with a mass spectrometer and a flame ionization detector (GC-MS/FID) at the supersites in Shanghai, Nanjing, and Hangzhou. A total of 55 PAMS (Photochemical Assessment Monitoring Stations) species were identified, including 27 alkanes, 11 alkenes, 1 acetylene, and 16 aromatics. Detailed descriptions of these sites are provided in the Supporting Information. CO2 concentrations were observed at an urban site named “Zhangjiang” and a background site named “Dongtan” in Shanghai. We use the difference in CO2 concentrations between the urban and background site (ΔCO2) to compare the changes of CO2 emissions. The location of CO2 monitoring sites and time series data are shown in Figure S3.

## RESULTS AND DISCUSSION

**Daily Emissions.** China has successively initiated lockdown measures since January 24, 2020. During the lockdown, the intercity traffic was strictly restricted except for some necessary supplies. The traffic in the city was also largely reduced due to the requirement of home isolation. Most factories and construction sites were suspended. Meanwhile, the 2020 SF holiday starting on January 24 was extended to February 9. Schools, commerce, and businesses were also suspended during this period. After the SF holiday, some production and business began to recover gradually. However, most people were still working in isolation at home. From the end of February to mid-March, cities have successively lifted lockdown measures, and most factories and enterprises have gradually entered a state of full operation. In view of this, we divide the first quarter of 2020 into four stages: Prelockdown (Pre, January 1–January 23, 2020), lockdown (LD, January 24–February 10, 2020), initial resumption of work (RS1, February 11–March 10, 2020), and full operation (RS2, March 11–March 31, 2020).

Figure 1 shows the evolution of air pollutant and CO2 emissions in the four stages in the YRD region as calculated by our approach. Emissions began to decline rapidly since mid-January 2020. During the LD stage, emissions all reached their minimum. The percentage decrease during the lockdown (E[Pre−LD]/Pre) reached a maximum of 31% (SO2), 53% (NO2),
34% (CO), 53% (NMVOCs), 35% (PM$_{2.5}$), 55% (BC), 37% (POC), and 40% (CO$_2$) for different pollutants. After entering the RS1 and RS2 stages, the emissions began to rise gradually, and basically returned to the Pre stage level at the end of March. In comparison, emissions in 2019 only declined slightly during the SF (February 5−11, 2019) and quickly returned to preholiday levels around mid-to-late February. The evolutions of emissions from each specific source are presented in Figure S4.

Regional Distribution. Figure 2 shows the spatial and temporal variations of NO$_x$, NMVOC, and CO$_2$ emissions and observational data. (a, e, i) Spatial distributions of NO$_x$, NMVOC, and CO$_2$ emission reduction ratios during LD stage relative to Pre stage (E$_{[Pre-LD]/Pre}$) in 12 km × 12 km resolution in the YRD region. The black cycles represent the concentration reduction ratios (C$_{[Pre-LD]/Pre}$) of ground-observed NO$_2$ and NMVOCs. (b−d) Comparisons of the daily variations of NO$_x$ emissions (black solid lines) and NO$_2$ concentrations (pink solid lines) in Shanghai (b), Jiangsu (c), and Zhejiang (d). The pink dashed lines represent the standard deviations. (f−h) Comparisons of the daily variations of NMVOC emissions and their concentrations in Shanghai (f), Nanjing (g), and Hangzhou (h). (j−l) Comparisons of the daily variations of CO$_2$ emissions in Shanghai (j), Jiangsu (k), and Zhejiang (l). Daily variations of CO$_2$ emissions in Shanghai are compared with the difference of CO$_2$ concentrations between urban and background sites. The observation data are all shown in 7-day moving averages. The daily variations of emissions and concentrations are all normalized based on the baseline emissions during the Pre stage.

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Figure 2. Spatial and temporal variations of NO$_x$, NMVOC, and CO$_2$ emissions and observational data. (a, e, i) Spatial distributions of NO$_x$, NMVOC, and CO$_2$ emission reduction ratios during LD stage relative to Pre stage (E$_{[Pre-LD]/Pre}$) in 12 km × 12 km resolution in the YRD region. The black cycles represent the concentration reduction ratios (C$_{[Pre-LD]/Pre}$) of ground-observed NO$_2$ and NMVOCs. (b−d) Comparisons of the daily variations of NO$_x$ emissions (black solid lines) and NO$_2$ concentrations (pink solid lines) in Shanghai (b), Jiangsu (c), and Zhejiang (d). The pink dashed lines represent the standard deviations. (f−h) Comparisons of the daily variations of NMVOC emissions and their concentrations in Shanghai (f), Nanjing (g), and Hangzhou (h). (j−l) Comparisons of the daily variations of CO$_2$ emissions in Shanghai (j), Jiangsu (k), and Zhejiang (l). Daily variations of CO$_2$ emissions in Shanghai are compared with the difference of CO$_2$ concentrations between urban and background sites. The observation data are all shown in 7-day moving averages. The daily variations of emissions and concentrations are all normalized based on the baseline emissions during the Pre stage.

Regional Distribution. Figure 2 shows the spatial and temporal variations of NO$_x$, NMVOC, and CO$_2$ emissions in the YRD region during the COVID-19 pandemic. Figure 2(a, e, and i) indicates that the E$_{[Pre-LD]/Pre}$ of NO$_x$, NMVOCs, CO$_2$ exhibit considerable differences in spatial variations. The NO$_x$ E$_{[Pre-LD]/Pre}$ in the southern area of the region is relatively higher (>80%), while the northern area was lower (<30%). The NMVOC E$_{[Pre-LD]/Pre}$ is higher (>70%) in the east and lower (<40%) in the west. The CO$_2$ E$_{[Pre-LD]/Pre}$ is higher (>60%) in the south and lower (<40%) in the north and west. The E$_{[Pre-LD]/Pre}$ values of other pollutants also show large spatial differences (Figure S5).

To verify the reliability of our estimated emission variations, we compare daily variations of NO$_x$, NMVOC, and CO$_2$ emissions with that of the observed NO$_x$, NMVOC, and ΔCO$_2$ concentrations. The 7-day moving average was used for comparison to minimize the interference caused by weather fluctuations. As shown in Figure 2, NO$_x$ and NMVOC emission variations in each province and city both have good agreements with the temporal variations of observation data, further demonstrating that our approach can accurately capture the temporal and spatial changes of air pollutant emissions. Considering that the above method cannot completely eliminate the interference of meteorological factors, we believe that this method is more suitable for short-lived precursors and for periods when emissions change drastically. Since CO$_2$ is a long-lived gas with high global background concentration, few studies have attempted to use observation data to verify its emission changes. As shown in Figure 2(j), the ΔCO$_2$ concentration and CO$_2$ emissions in Shanghai simultaneously show declines of the same magnitude at the beginning of LD stage and maintain the same magnitude of changes during the RS1 and RS2 stages, which suggests that ΔCO$_2$ concentrations can be used to characterize the CO$_2$ emission changes. This finding points to a new approach for evaluating the anthropogenic CO$_2$ emissions through well designed monitoring network.
Contribution of Individual Sources and Synergy. To distinguish the impacts of COVID-19 lockdowns from the SF effect, we estimate the daily emission differences of air pollutants and CO₂ during the same lunar calendar days between 2019 and 2020. Figure 3 shows the contribution of each individual source to the emission differences for major air pollutants and CO₂. The reductions of SO₂ emissions were mainly attributed to power plants, cement manufacturing, and textile sectors, respectively reduced by 3.1%, 8.0%, and 3.3% relative to the total emissions during the same period in 2019. NOₓ emission reductions were strongly dominated by Mobile, especially for diesel and gasoline vehicles, contributing 8.6% and 3.8% of the total emission differences due to the COVID-19 lockdowns. Recent studies have also attributed the NOₓ reduction during the COVID-19 pandemic to vehicles. However, it should be noted that the contributions of the power plants and cement manufacturing sectors to the NOₓ reduction were still not negligible, contributing 2.5% and 4.3% of emission reductions, respectively. NMVOC emission reduction mainly came from gasoline vehicles (7.5%) and major NMVOC-related industries, like the industrial coating (7.2%), rubber and plastic (3.5%), and chemical manufacturing (2.2%) sectors, as well as oil storage and transportation sector (3.1%). The main sources contributing the CO₂ emission reductions were power plants and cement manufacturing sector, which have reduced total emissions by 6.7% and 8.1%.

Figure 3. Contributions of individual sources to total emission differences of air pollutants and CO₂ between 2019 and 2020. (a−d) Ration differences in SO₂ (a), NOₓ (b), NMVOCs (c), and CO₂ (d) emissions contributed by each individual source during the same lunar calendar days. Data are aligned according to the Chinese lunar calendar. The x-axis represents the lunar day. (e) Emissions reduction rates of each individual source during this period.
respectively. Gasoline and diesel vehicles also contributed 3.9% and 1.2% of CO2 emission reductions, respectively. Figure 3(e) compares the synergy of COVID-19 lockdowns on major air pollutants and CO2 emissions from each individual source. Overall, SO2 and NOx reductions both show synergy with CO2 reductions in Power, H-Ind, and L-Ind sectors. This is mainly due to the strong homology of SO2, NOx, and CO2 emissions in these sectors, which account for 58%, 34%, and 70% of total SO2, NOx, and CO2 emissions in the YRD region, respectively (Figure S7). Besides the sectors mentioned above, NOx reductions in Mobile have stronger synergy with CO2 reductions. Especially for gasoline vehicles, their declines have the same contributions to total reductions of NOx and CO2 emissions in the YRD region, while the declines of diesel vehicles have more contributions on NOx reduction. Comparatively, the synergy between NMVOCs and CO2 emission reductions is relatively poor. Except for gasoline vehicles, there is almost no synergy between NMVOCs and CO2 reductions in other sectors. Our results indicate that there are considerable differences in the synergy of air pollutants and CO2 emission reductions for various sources. Future emission reduction policies need to develop more refined control strategies for different source categories according to their synergy. For example, the Power and H-Ind sectors need to explore more structural adjustment measures in the future to tapping the reduction potential of CO2 emissions while reducing SO2 and NOx emissions. For gasoline vehicles, promoting their fleet electrification will simultaneously reduce NOx, NMVOCs, and CO2 emissions and have a synergistic effect on PM2.5 and ozone pollution control and greenhouse gas mitigation, while for diesel vehicles more efforts should be engaged to reduce their NOx emission levels. Future reductions of NMVOC emissions should focus more on the governance of NMVOCs-related sectors, such as chemical manufacturing, textile, industrial and residential coating, and others.

This study constructed a city-level high resolution dynamic emission inventory for air pollutants and CO2 by a dynamic temporal allocation coefficient driven approach and first applied it in the YRD region, China. The uncertainties in our results mainly lie in the determination of dynamic temporal allocation coefficients. For example, we used CEMS data to “point-to-point” drive the daily variations of emissions from large point sources. However, the CEMS system only monitored the organized emissions. For the unorganized emissions, we assumed their variations were consistent with the organized. In addition, CEMS only monitored SO2, NOx, and PM emissions; other pollutants were substituted by SO2 and PM as proxies. For NMVOC emissions, we used IEC data instead to estimate their variations. This proxy method will introduce large uncertainties. However, the COVID-19 lockdown provides a good opportunity to test the approach. The comparison between emission estimates and observation data shows that our approach can well capture the spatiotemporal changes of both short-lived precursors (NOx and NMVOCs) and CO2 emissions. This approach is potentially applicable to other regions where real-time multisource activity data are available. It can also be applied to the scientific assessment of short-term emission control measures during societal events and long-term changes of emissions.

China’s air pollution prevention and control measures have paid more attention to end-of-pipe control reductions in these years, which is effective in reducing SO2, NOx, and primary PMx emissions but cannot achieve coordinate control of air pollutants and CO2 emissions. Recent studies suggest the benefits of end-of-pipe control reductions will be exhausted by 2030, and further improvement of air quality must rely on more ambitious low-carbon policies. The unintentional emission reduction case caused by the COVID-19 pandemic indicates that we should implement differentiated control strategies for different source categories. For example, coal-fired power plants in China have reduced 65% and 60% of SO2 and NOx emissions by implementing a ultralow emission (ULE) standard between 2013 and 2017, and the industrial SO2 and NOx emissions have also been reduced by 62% and 11%, while their CO2 emissions are still on the rise. The COVID-19 lockdown case indicates that the reductions of CO2 in Power and H-Ind sectors caused by the declines in activity level have synergistic effects on SO2 and NOx reductions. From a long-term perspective, these sectors should rely more on energy and industrial structure adjustment measures to achieve coordinated reduction goals. Accelerating the fleet electrification of on-road vehicles can contribute to the simultaneous reductions of NOx, NMVOCs, and CO2 emissions. In addition, we still need to step up efforts to control key sources that have important contributions to a single kind of pollutants, like NMVOC emissions from L-Ind sectors and NOx emissions from diesel vehicles, which is crucial to improving air quality at the current stage.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.1c00600. Tables S1−S4 (XLSX) Description and Figures S1−S12 (PDF)

AUTHOR INFORMATION

Corresponding Authors

Hang Su – State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China; Max Planck Institute for Chemistry, Mainz 55128, Germany; orcid.org/0000-0003-4889-1669; Email: h.su@mpic.de

Cheng Huang – State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China; orcid.org/0000-0001-9518-3628; Email: huangc@saes.sh.cn

Hongli Wang – State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China; orcid.org/0000-0003-0655-3389; Email: wanghl@saes.sh.cn

Authors

Jingyu An – State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China

Qizhen Liu – Shanghai Environmental Monitoring Centre, Shanghai 200235, China
Junjie Tian — State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China

Qian Wang — State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China

Qingyao Hu — State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China

Rusha Yan — State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex, Shanghai Academy of Environmental Sciences, Shanghai 200233, China

Yin Shen — Shanghai Environmental Monitoring Centre, Shanghai 200235, China

Yusen Duan — Shanghai Environmental Monitoring Centre, Shanghai 200235, China

Qingyan Fu — Shanghai Environmental Monitoring Centre, Shanghai 200235, China

Jiadong Shen — Hangzhou Ecological Environment Monitoring Center of Zhejiang Province, Hangzhou 310007, China

Hui Ye — Hangzhou Ecological Environment Monitoring Center of Zhejiang Province, Hangzhou 310007, China

Ming Wang — Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing 210044, China

Chong Wei — Shanghai Carbon Data Research Center, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201210, China; orcid.org/0000-0003-0632-4324

Yafang Cheng — Max Planck Institute for Chemistry, Mainz SS128, Germany; orcid.org/0000-0003-4912-9879

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.estlett.1c00600

Notes

The authors declare no competing financial interest.

Data Availability Statement. Data used in this paper can be found online at https://data.mendeley.com/datasets/92mp3bbxpy/1.

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