



**Rapid assessment of drivers and air quality effects of  
regional daily changes in air pollutant emissions based on  
near-real-time techniques: A case in Jiangsu Province, China**

Chen Gu<sup>1</sup>, Yutong Wang<sup>1,5</sup>, Yuan Ji<sup>1</sup>, Lei Zhang<sup>1,2</sup>, Shuanzhu Sun<sup>3</sup>, Yuandong Bian<sup>1</sup>,  
Zimeng Zhang<sup>1</sup>, Jiewen Zhu<sup>3</sup>, Wenxin Zhao<sup>1</sup>, Sheng Zhong<sup>4</sup>, Yu Zhao<sup>1,2\*</sup>

<sup>1</sup> State Key Laboratory of Water Pollution Control and Green Resource Recycling and  
School of Environment, Nanjing University, 163 Xianlin Rd., Nanjing, Jiangsu  
210023, China

<sup>2</sup> Collaborative Innovation Center of Atmospheric Environment and Equipment  
Technology, CICAET, Nanjing, Jiangsu 210044, China

<sup>3</sup> Jiangsu Frontier Electric Power Technology Co., Ltd., 58 Suyuan Ave., Nanjing,  
Jiangsu 211102, China

<sup>4</sup> Jiangsu Provincial Environmental Monitoring Center, 100 Zhonghe Rd., Nanjing  
210013, China

<sup>5</sup> Key Laboratory of Formation and Prevention of Urban Air Pollution Complex,  
Ministry of Ecology and Environment, Shanghai Academy of Environment Sciences,  
Shanghai 200233, P. R. China

\*Corresponding author: Yu Zhao

Phone: 86-25-89680650; Email: yuzhao@nju.edu.cn



## 27 ABSTRACT

28 Fast and timely estimation of changing air pollutant emissions is critical for  
29 understanding the complex sources of air pollution and supporting air quality  
30 improvement, while current regional emission inventory was commonly reported with  
31 time lag or coarse temporal resolution. Here we developed a near-real-time approach  
32 that calculates the daily emissions of anthropogenic air pollutants, and applied this  
33 approach for Jiangsu province, a typical developed region in eastern China. We  
34 estimated that the annual total anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, primary fine  
35 particles (PM<sub>2.5</sub>), non-methane volatile organic compounds (NMVOCs), and NH<sub>3</sub>  
36 were 246, 727, 298, 1186, and 377 Gg, respectively, for Jiangsu in 2022. Compared to  
37 the national emission inventory, application of the provincial-level daily emission  
38 estimates provided better model performance of PM<sub>2.5</sub> and ozone (O<sub>3</sub>) simulation for  
39 all the involved months. The NO<sub>x</sub>, SO<sub>2</sub>, PM<sub>2.5</sub>, and NMVOCs emissions in Jiangsu  
40 during April-May 2022 (the period of COVID-19 lockdown in Shanghai) were  
41 respectively 8%, 6%, 6%, and 10% smaller than those in the same period of 2023.  
42 Transportation and Industry respectively contributed 89% of NO<sub>x</sub> emission reduction  
43 and 93% NMVOCs reduction. Combining with machine learning algorithms,  
44 moreover, we revealed that the changing agricultural NH<sub>3</sub> emissions dominated the  
45 variability of daily PM<sub>2.5</sub> concentration, and that off-road transportation contributed  
46 substantially to variabilities of both PM<sub>2.5</sub> and O<sub>3</sub> levels. The study proved advantages  
47 of incorporation of near-real-time data and machine learning techniques on tracking  
48 the fast-changing emissions and detecting the sources of varying air quality.  
49



## 50 1. Introduction

51 Emissions of air pollutants from anthropogenic activity including traffic, industrial  
52 plants, and residential and commercial fuel consumption are the main cause of  
53 worsened air quality, especially in economically developed regions with dense  
54 populations (Sokhi et al., 2022; Zheng et al., 2018). Emission inventory, which  
55 contains complete information on magnitude, spatial pattern, and temporal change of  
56 air pollutant emissions by sector, is essential for identifying the sources of air  
57 pollution and effectiveness of emission controls on air quality through numerical  
58 modeling (Zhao et al., 2013; Zhang et al., 2019). Traditionally, “bottom-up”  
59 methodology (i.e., the emissions were calculated for the finest source categories and  
60 then aggregated to bigger categories) provides robust time series of emission  
61 estimates based on national statistics (An et al., 2021; Crippa et al., 2020; Kurokawa  
62 et al., 2020). However, these emission estimates were usually reported with a time lag  
63 of at least 3-5 years. The delay reflected the time needed to finalize accurate national  
64 statistics (e.g., official energy consumption by fuel type) and that needed to collect  
65 and process them for compiling emission inventories (Guevara et al., 2023). As a  
66 result, in addition to the inherent uncertainties in emission inventories, this delay can  
67 introduce extra uncertainty when these inventories are employed in air quality  
68 modeling, as they may miss current emission characteristics (Tong et al., 2012). Such  
69 limitation can be greatly exacerbated for periods with big and unexpected emission  
70 fluctuations, resulting from temporary actions for major events or public health  
71 incidents (Huang et al., 2021; Wang et al., 2025).

72 To better track the changing emissions for specific events or incidents (e.g.,  
73 COVID-19 pandemic), researchers have developed alternative methods to obtain the  
74 near-real-time emission estimates (Gaubert et al., 2021; Schneider et al., 2022). The  
75 objective of these efforts is to understand the driving factors of the changing  
76 emissions and their impact on air quality. Real-time activity information with high  
77 temporal resolution started to be incorporated in the emission estimation, such as the



78 electricity load and generation data by national transmission system operators, the  
79 real-time vehicle flows monitored from navigation applications, and the real-time ship  
80 navigational information from automatic identification system (AIS) (Liu et al., 2020a;  
81 Liu et al., 2020b; Zheng et al., 2021; Huang et al., 2021; Harkins et al., 2021; Guevara  
82 et al., 2021). Although limited availability and huge capacity of these data hinder their  
83 full use in emission inventory development, there is a big potential in expanding the  
84 data source to improve the capability of capturing the fast-changing emissions.

85 Currently, studies have been conducted for carbon dioxides (CO<sub>2</sub>) emissions and  
86 near-real-time data platforms and products have been developed, particularly for  
87 well-identified stationary sources such as fossil fuel combustion plants (BEIS, 2022;  
88 CBS, 2024; CITEPA, 2024; Carbon Monitor, 2024). Comparatively, achieving  
89 near-real-time estimates is more challenging for air pollutants due to the large  
90 complexity and variability of their emission processes. A great variety of air pollutants  
91 come from a wide range of sources, containing fuel combustion, industrial processes,  
92 on-road and off-road traffic, solvent evaporation, and agricultural activities (Xu et al.,  
93 2023; Zheng et al., 2020). The emissions can be greatly influenced by many factors  
94 and change a lot. Those factors include the human behavior patterns, operating  
95 conditions of plants, improved use of manufacturing and pollution control  
96 technologies, and/or meteorological conditions (Liu et al., 2024; Lei et al., 2023;  
97 Geng et al., 2024). Given the strong chemical reactivity and short atmospheric  
98 lifetime of many air pollutants, there exist complicated relationships between  
99 emissions and air quality, emphasizing the importance of tracking the fast-changing  
100 emissions (Liu et al., 2020; Zhao et al., 2020a). Therefore, efforts are still in great  
101 need to develop effective approach for estimating the near-real -time emissions.

102 For the past years, China has substantially enhanced emission control for industrial  
103 (e.g., “ultra-low” emission retrofit for selected non-electrical industries) and  
104 residential sources (e.g., promotion of advanced stoves and clean coals during heating  
105 seasons). Those measures have clearly reduced emissions of many air pollutants,  
106 resulting in a 17.2 µg/m<sup>3</sup> decline of fine particle (PM<sub>2.5</sub>) concentration between 2015



107 and 2020 over the country (Geng et al., 2024). In contrast, the emissions of  $\text{NO}_x$  and  
108  $\text{PM}_{2.5}$  from passenger transportation respectively grew by 178% and 152% from 2019  
109 to 2022 (Zhang et al., 2023), and the maximum daily 8h mean ozone (MDA8  $\text{O}_3$ )  
110 concentrations increased 5.8% from 2021 to 2022 for the country (MEE, 2023). The  
111 diverse changes in emissions and air quality highlight the necessity to quickly and  
112 accurately reveal the drivers of changes in air pollutant emissions and their impact on  
113 ambient air quality (Gu et al., 2023). This is particularly important for periods with  
114 severe air pollution episodes and unexpected incidents that substantially changed  
115 human activities like COVID-19 lockdown, as timely temporary actions to address  
116 pollution might be urgently required.

117 Province serves as a crucial role in air quality management in China. Due to  
118 difference in economic and energy structure and atmospheric conditions, local  
119 governments often implement diverse strategies and actions to reduce regional air  
120 pollution. This results in large variability in both emission and air quality changes  
121 across different regions. (Liu et al., 2022; Wang et al., 2021). Studies relying on  
122 national emission data offer limited guidance in developing emission control  
123 measures and assessing their effectiveness in air quality improvement (An et al.,  
124 2021). Jiangsu Province, located in the Yangtze River Delta (YRD) in eastern China,  
125 is one of most economically developed regions across the country (Supplementary  
126 Figure S1). It accounted for 10.2% of the gross domestic product (GDP) in mainland  
127 China (ranking the second place in the country), and 8.1%, 12.4% and 11.6% of coal  
128 consumption, cement and crude steel production in 2022, respectively (NBS, 2023).  
129 Following the implementation of air pollution prevention measures, the  $\text{PM}_{2.5}$   
130 pollution in Jiangsu has significantly decreased since 2015. However, the  
131 development of the petrochemical industry and transportation has led to rapid changes  
132 in emissions, making Jiangsu as the province with the highest and fastest growing  $\text{O}_3$   
133 concentration in YRD in recent years (Zhou et al., 2017; Wang et al., 2022).

134 In this study, therefore, we selected Jiangsu as an example to demonstrate the  
135 development of near-real-time emission inventory and its application on rapid



assessment of air quality. Based on our previous work that incorporated the best available facility-level information to develop a comprehensive provincial emission inventory (Gu et al., 2023), here we constructed an approach driven by real-time activity data from multiple sources. The pollutants include  $\text{SO}_2$ ,  $\text{NO}_x$ , primary  $\text{PM}_{2.5}$ ,  $\text{NH}_3$ , and non-methane volatile organic compounds (NMVOCs). We then applied the method to obtain the near-real-time emission estimates for 2022-2023, and assessed the driving factors of the short-term emission change during the COVID-19 lockdown period. Finally, we used an Extreme Gradient Boosting (XGBoost) algorithm to explore the relationship between the variability of daily  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentrations and their precursor emissions for 2022. The study provides insights for timely design and implementation of air pollution control actions, and can be used for reference for other developed and polluted regions in China and worldwide.

## 2. Methodology and data

### 2.1 Framework of near-real-time emission estimation

Figure 1 shows the methodological framework. In our previous study (Gu et al., 2023), we collected, examined, and integrated most available information on emission sources to enhance the completeness and reliability of the provincial emission inventory. All the information, including raw material and energy consumption, product output, and manufacturing and emission control technologies, played an important role in the estimation of near real-time emissions. The specific methods by sector are described in Section 2.2. Furthermore, we improved the spatial distribution of air pollutant emissions. Point sources of power and industrial enterprises were allocated based on their precise latitudes and longitudes. We further utilized Point of Interest (POI) data from Gaode Map (<https://lbs.amap.com/>, last visited on October 2025) to obtain the real-time changes on road and waterway networks, land use, and building footprints. The information is updated every 2-3 months. The use of POI data significantly reduced the error of spatial allocation of emissions that may result from the delayed and indirect information on the “surrogate” parameters (Wang et al.,



164 2017).

## 165 2.2 Near-real-time daily emission estimation by sector

166 This section describes the methods for estimating near-real-time daily emissions for  
 167 2022 and 2023. Six major sectors were included (Power, Industrial plant, Vehicles  
 168 (On-road transportation), Off-road machinery, Residential, and Agriculture), covering  
 169 most anthropogenic activities. Road and construction site dusts were not contained.

170 **Power plant** Previously we developed a method of applying online measurement data  
 171 from the continuous emission monitoring systems (CEMS,  
 172 <http://218.94.78.61:8080/newPub/web/home.htm>, last visited on October 2025) for  
 173 emission estimation at the unit/plant level (Zhang et al., 2019). With this basis, we  
 174 have improved the emission estimation method to enable the stable and continuous  
 175 acquisition of near-real-time emission data lagged by one month. For the small  
 176 number of power-generating units without CEMS data, we assumed that their  
 177 pollutant concentrations in the flue gas were at the average level of units with similar  
 178 installed capacity (Tang et al., 2019). The emissions were calculated based on the  
 179 mean hourly flue gas concentration of air pollutant obtained from CEMS and the  
 180 theoretical flue gas volume of each unit/plant:

$$181 \quad E_{i,j,day} = C_{i,j,month} \times AL_{j,month} \times V_{j,m}^0 \times P_{i,j,m,day} \quad (1)$$

$$182 \quad AL_j = F_m / R_m \quad (2)$$

183 where  $E$  is the emission of air pollutant;  $i, j$  and  $m$  indicate the specific pollutant  
 184 species, individual power plant or unit, and fuel type, respectively;  $C$  is the monthly  
 185 average concentration in the flue gas;  $AL$  is the activity level (here monthly coal  
 186 consumption);  $F$  is the monthly electricity generation for various fuels, as reported by  
 187 NBS (2023);  $R$  is the fuel consumption rate for power generation, taken from Tong et  
 188 al. (2021),  $V^0$  is the theoretical volume of flue gas produced per unit of fuel  
 189 consumption (Zhao et al., 2010);  $P$  is the temporal profile of emissions (the daily to  
 190 monthly emission ratio), based on the hourly pollutant concentrations and volume of  
 191 flue gas for the month and specific day.



192 **Industrial plant** With its gradually expanding penetration, CEMS has become able to  
193 support near-real-time emission estimation for industrial plants (Tang et al., 2022; Bo  
194 et al., 2021). Given its varying coverage across sectors, we have developed a method  
195 that can stably estimate the near-real-time emissions at the plant level with a lag of  
196 one month. This method classifies industrial plants into three categories based on their  
197 CEMS coverage, as described below.

198 (1) Industrial plants with CEMS information. The method is similar to power plants:

$$199 \quad E_{i,j,day} = C_{i,j,month} \times AL_{j,month} \times V_{i,j,k}^0 \times P_{i,j,m,day} \quad (3)$$

200 where  $k$  denotes the industrial sector;  $AL$  is the activity level (here represents monthly  
201 product output) as reported by NBS (2023), and  $V^0$  is the theoretical volume of flue  
202 gas produced per unit of product output, which can be found in the technical  
203 specifications for the application of emission permits (MEE, 2021).

204 (2) Industrial plants without CEMS while it was equipped at some plants within the  
205 same sector. Sector-level emission factors (emissions per unit of activity level, EF)  
206 were calculated using CEMS data from other plants. Monthly emissions were  
207 estimated based on the sector-level EF and monthly product output from official  
208 environmental statistics. The near-real-time daily emissions were then generated  
209 according to the temporal profile of emissions ( $P$ ) obtained from CEMS installed in  
210 other available plants in the sector.

$$211 \quad E_{i,j,day} = AL_{j,month} \times EF_{i,k} \times P_{i,j,m,day} \quad (4)$$

$$212 \quad EF_{i,k} = E_{i,k,month} / AL_{k,month} \quad (5)$$

213 where  $EF_{i,k}$  is the sector-average emission factor for plants with CEMS for sector  $k$ ,  
214  $E_{i,k}$  and  $AL_k$  are the total emissions from industrial plants with CEMS and their  
215 product output, respectively.

216 (3) Industrial sectors without CEMS data. Emissions were principally calculated  
217 based on activity level and emission factor. The activity data were derived based on  
218 monthly official statistics reported by NBS (2023). In addition, we analyzed the  
219 historical emission source data to trace the evolution of manufacturing and emission  
220 control technologies for various sectors, and the emission factors could be calculated





221 for near-real-time emission estimations:

$$222 \quad E_{i,day} = AL_{month} \times EF_{i,k} \times P_{i,m,day} \quad (6)$$

223 where  $EF$  represents the emission factor based on the technological evolution of the  
224 plant,  $P$  is the temporal profile of emissions, based on the fraction of daily electricity  
225 load out of the monthly total for specific sector.

226 **Vehicles (On-road transportation)** Daily vehicular emissions were estimated  
227 utilizing the International Vehicle Emissions model (IVE) combined with the Gaode  
228 live congestion index (Zhou et al., 2019; Kholod et al., 2016). The level of traffic  
229 congestion was indicated by the additional time incurred during a trip under congested  
230 conditions, expressed as a percentage relative to uncongested conditions (Huo et al.,  
231 2022). The Gaode congestion index is available for over 350 cities in China, with a  
232 temporal resolution of 5 minutes (<https://report.amap.com/index.do>, last visited on  
233 October 2025). By integrating the congestion index with a Greenshield's traffic  
234 density model (Yang et al., 2019), we estimated the traffic volume which serves as a  
235 temporal allocation factor to calculate the daily emissions. This approach assumes that  
236 vehicular activity data (e.g., mileage and fuel consumption) are accessible, albeit  
237 typically with a lag in reporting, as such information is usually provided on an annual  
238 basis. Consequently, the near-real-time emissions can be estimated based on the daily  
239 variations of congested index and EFs compared to the previous year (Eq. 7):

$$240 \quad E_{i,m,day} = \frac{(I_{day,year-1}) \times I_{day,(year-1)}^2 \times EF_{i,m,day,year}}{(I_{day,(year-1)} - 1) \times I_{day,year}^2 \times EF_{i,m,day,(year-1)}} \quad (7)$$

241 where  $I$  is the Gaode traffic congestion index; and  $EF$  is the emission factor,  
242 calculated by the IVE model. The input parameters of IVE such as the vehicle  
243 population by type, registration dates, fuel types, and emission standards, can be  
244 obtained from the transportation management departments of individual cities. These  
245 historical data can be extrapolated to the present date utilizing the vehicle survival  
246 curve, thereby bridging any gaps in the current information (Sun et al., 2020).

247 **Off-road Transportation** Off-road transportation was divided into five categories:  
248 construction machinery, agricultural machinery, marine, railway, and aviation.  
249 Emissions from construction machinery were estimated based on assumed daily



250 utilization rates derived from the operating rates of construction sites (Shen et al.,  
 251 2023; Huang et al., 2021). The daily usage of agricultural machinery was assumed to  
 252 correlate with the application of nitrogen fertilizers from agricultural sources (see the  
 253 description of agriculture as below). Emissions from railway, marine and aviation  
 254 sources were estimated using data from passenger/cargo turnover, individual ports and  
 255 commercial flights, respectively. These data were obtained from the China  
 256 Entrepreneur Investment Club (CEIC) (<https://www.ceicdata.com.cn>, last visited on  
 257 October 2025), Marine Traffic (<http://www.marinetraffic.com>, last visited on October  
 258 2025) and Flightradar24 databases (<http://www.flightradar24.com>, last visited on  
 259 October 2025) (Huo et al., 2022; Liu et al., 2020a).

260 **Residential sources** We followed Shao et al. (2023) and developed a Bayesian  
 261 hierarchical model to estimate daily heating energy consumption by fuel type, based  
 262 on two primary factors influencing residential energy consumption: temperature and  
 263 GDP. The daily temperature data were taken from ERA5 products provided by the  
 264 European Centre for Medium-Range Weather Forecasts (ECMWF)  
 265 (<https://cds.climate.copernicus.eu>, last visited on October 2025), while GDP from the  
 266 national statistics published quarterly by the National Bureau of Statistics  
 267 (<http://www.stats.gov.cn/>, last visited on October 2025). For the months without GDP  
 268 data, we assumed a linear relationship between GDP and the nighttime light index (Xu  
 269 et al., 2024), and applied the National Polar-orbiting Partnership Visible Infrared  
 270 Imaging Radiometer Suite (NPP-VIIRS, <https://www.earthdata.nasa.gov/>, last visited  
 271 on October 2025) provided by National Aeronautics and Space Administration  
 272 (NASA) to extrapolate the GDP for those months. We applied the gridded population  
 273 dataset (1km×1km) released by a database of the Chinese Academy of Sciences  
 274 (<https://www.resdc.cn/Default.aspx>, last visited on October 2025) for 2020. To  
 275 account for the effect of large-scale population migration, we integrated the  
 276 Population Migration Index (PMI) developed by Baidu (<https://qianxi.baidu.com/#/>,  
 277 last visited on October 2025). This index calculates the proportion of incoming  
 278 migrants relative to the local population.



279 **Agriculture**  $\text{NH}_3$  emissions from fertilizer use can be largely influenced by  
280 meteorological conditions, soil environment, and farming practices. In our previous  
281 study, we quantified  $\text{NH}_3$  emissions using dynamic EFs associated with those factors  
282 (Zhao et al., 2020b). In this study, we expanded the methodology and estimated  $\text{NH}_3$   
283 emissions by using daily EFs. For livestock and poultry farming, we assumed that  
284 daily  $\text{NH}_3$  emissions were associated with temperature, while those for human  
285 excretion associated with both temperature and PMI.

### 286 **2.3 Air quality modeling**

287 To evaluate the near-real-time emission estimate, we used the Community Multiscale  
288 Air Quality (CMAQ v5.1) model developed by US Environmental Protection Agency  
289 (<https://www.epa.gov/cmaq>, last visited on October 2025), to simulate the  $\text{PM}_{2.5}$  and  
290  $\text{O}_3$  concentrations in Jiangsu. Four months (January, April, July, and October) in 2022  
291 were selected as the simulation periods, with a spin-up time of 7 days for each month  
292 to reduce the impact of the initial condition on the simulation. As shown in  
293 Supplementary Figure S1, three nested domains (D1, D2, and D3) were applied with  
294 the horizontal resolutions at 27, 9, and 3 km, respectively, and the most inner D3  
295 covered Jiangsu and parts of the YRD region including Shanghai, northern Zhejiang,  
296 and eastern Anhui. The Multi-resolution emission inventory of China (MEIC, [http://](http://meicmodel.org.cn/)  
297 <http://meicmodel.org.cn/>, last visited on October 2025) was applied for D1, D2, and  
298 the regions out of Jiangsu in D3 (Zheng et al., 2018), and the provincial-level  
299 near-real-time emission estimate was applied for Jiangsu in D3. The Carbon Bond  
300 Mechanism (CB05) and AERO5 mechanisms were used for the gas-phase chemistry  
301 and aerosol module, respectively.

302 The meteorological field for the CMAQ was obtained from the Weather Research and  
303 Forecasting model (WRF v3.4, <https://www.mmm.ucar.edu/models/wrf>, last visited on  
304 October 2025). Meteorological initial and boundary conditions were obtained from  
305 the National Centers for Environmental Prediction (NCEP,  
306 <https://psl.noaa.gov/data/reanalysis/reanalysis.shtml>, last visited on October 2025)  
307 datasets. Ground observations at 3-h intervals were downloaded from National



308 Climatic Data Center (NCDC, <ftp://ftp.ncdc.noaa.gov/pub/data/noaa/isd-lite/>, last  
 309 visited on October 2025). Statistical indicators including bias, index of agreement  
 310 (IOA), and root mean squared error (RMSE) were used to evaluate the WRF  
 311 performance (Gu et al., 2023). The discrepancies between simulations and ground  
 312 observations were within an acceptable range (Supplementary Table S1).  
 313 We collected ground observation data of hourly  $PM_{2.5}$  and  $O_3$  concentrations at the  
 314 110 state-operating air quality monitoring stations within Jiangsu  
 315 (<https://data.epmap.org/page/index>, see the station locations in Figure S1, last visited  
 316 on October 2025). Correlation coefficients (R), normalized mean bias (NMB) and  
 317 normalized mean errors (NME) between observation and simulation for each month  
 318 were calculated to evaluate the performance of CMAQ modeling.  
 319 We further compared the modeling performance using provincial-level emission  
 320 estimate in D3 with that using MEIC. MEIC was currently available only for 2020. To  
 321 avoid the bias from the total emission level, we adjusted the total emissions of various  
 322 species in 2020 MEIC to be consistent with our estimates, retaining the  
 323 spatiotemporal and sector distribution of the emissions.

#### 324 **2.4 Removing meteorological influence on $PM_{2.5}$ and $O_3$ concentrations**

325 To explore the influence of anthropogenic emission changes on the variability of  
 326  $PM_{2.5}$  and  $O_3$  levels in 2022, we removed the impact of varying meteorological  
 327 conditions by employing a stepwise multiple linear regression (MLR) model (Li et al.,  
 328 2021). The surface daily concentrations of  $O_3$  and  $PM_{2.5}$  were taken from the Tracking  
 329 Air Pollution in China (TAP, <http://tapdata.org.cn/>, last visited on October 2025) with  
 330 a horizontal resolution of 1 km×1 km (Geng et al., 2021). We incorporated nine  
 331 meteorological variables from the ERA5 database at a resolution of 0.25°×0.25°,  
 332 considered as the potential covariates for  $O_3$  and  $PM_{2.5}$ . They were 10-meter zonal and  
 333 meridional wind speeds, temperature, boundary layer height, sea level pressure, cloud  
 334 cover, precipitation, relative humidity, and dew point temperature. These variables  
 335 were then scaled to a 3km×3km grid system by bilinear interpolation. To prevent  
 336 overfitting, we conducted MLR with the three most influential meteorological



parameters to estimate the variability of daily  $PM_{2.5}$  and maximum daily 8-hour average (MDA8)  $O_3$  concentration for each grid cell. Anomaly (the difference between the raw data and the moving average of 30 days around) of air pollutant concentrations and meteorological factors were used in the model, to exclude the effect of monthly variability. Residuals that cannot be explained by the meteorological variables were assumed to be attributed to anthropogenic emission changes (Li et al., 2020). The results could be interpreted as the sensitivity of air pollutant concentration to the daily emission anomalies from the annual average value.

To evaluate the MLR performance, we collected daily  $PM_{2.5}$  and  $O_3$  concentrations at the above-mentioned 110 air quality monitoring stations in Jiangsu (Figure S1), and the R and NMB between observation and MLR were calculated.

## **2.5 Examining the response of MDA8 $O_3$ and $PM_{2.5}$ concentration to changing daily emissions**

### **2.5.1 XGBoost model**

XGBoost model is an advanced and scalable machine learning framework based on gradient-boosted decision trees, widely recognized for its efficiency in handling structured data and modeling complex nonlinear relationships (Requia et al., 2020; Wang et al., 2023). XGBoost excels at processing high-dimensional spatiotemporal datasets, such as gridded emission inventories, by effectively capturing interactions among heterogeneous emission sources and temporal dependencies. Moreover, the inherent interpretability features facilitate seamless integration with explainable AI tools (e.g., SHapley Additive exPlanations (SHAP) to quantify the marginal contribution of each input feature to individual model predictions), enabling rigorous attribution analysis of air pollutant concentration variability (Zhao et al., 2025). The SHAP value is calculated with following equation:

$$y_i = y_{base} + f(X_{i,1}) + f(X_{i,2}) + \cdots f(X_{i,n}) \quad (8)$$

where  $y_i$  is the predicted value of the model for the  $i$ th sample;  $f(X_{i,n})$  is the



364 contribution of the  $n$ th eigenvalue in the  $i$ th sample to the final predicted value, with  
365 positive or negative representing that the eigenvalue makes the predicted value  
366 increase or decrease; and  $y_{base}$  is the baseline value of the predicted outputs for all  
367 types of predictions, representing the average prediction results for each category  
368 without the influence of any eigenvalue.

### 369 **2.5.2 Anthropogenic effects on PM<sub>2.5</sub> and MDA8 O<sub>3</sub> variability**

370 The XGBoost-SHAP modeling framework was implemented at the horizontal  
371 resolution of 3km×3km to capture the emission-concentration relationship. XGBoost  
372 regression models were independently trained for each grid cell. January and July  
373 were selected as typical months for ambient PM<sub>2.5</sub> and O<sub>3</sub>, respectively. Daily time  
374 series of 20 pollutant-sector combinations (4 pollutant (SO<sub>2</sub>, NO<sub>x</sub>, NMVOCs, PM<sub>2.5</sub>)  
375 × 5 sectors (Power, Industry, On-road (Vehicles), Off-road, Residential) except for  
376 tiny On-road SO<sub>2</sub>, and agricultural NH<sub>3</sub>,) were set as predictors, and  
377 anthropogenic-driven variability of PM<sub>2.5</sub> or O<sub>3</sub> concentrations as target variables.  
378 Similarly, the emission inputs were treated as anomaly (the difference between the  
379 current day's emissions and the moving average of 30 days around). A 10-fold  
380 cross-validation was applied (80% training and 20% testing), and the bias and  
381 correlation coefficient (R) were calculated to evaluate the model performance (Xiao et  
382 al., 2018).

383 SHAP values were calculated for each emission feature using the tree explainer  
384 algorithm, quantifying contributions of pollutant-sector combinations to variability of  
385 daily anthropogenic-driven concentrations. Note that SHAP values represented the  
386 deviation of individual predictions from the baseline expectation. Positive values  
387 indicated emission features that elevated pollutant concentrations above the baseline,  
388 while negative values indicated features that reduced concentrations below the  
389 baseline. Aggregation of daily SHAP values for various pollutant-sector combinations  
390 produced the daily-level contribution of total anthropogenic emissions to the changing  
391 ambient concentration, and the daily-level contributions could then be aggregated to



the monthly level.

### 3. Results and discussions

#### 3.1 Anthropogenic air pollutant emissions

##### 3.1.1 Total air pollutant emissions in 2022

The total anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, NMVOCs, and NH<sub>3</sub> in Jiangsu for 2022 were estimated at 246, 727, 298, 1186, and 377 Gg (Supplementary Figure S2), which were respectively reduced by 17%, 33%, 18%, 7%, and 11% compared with those in 2019 (Gu et al., 2023). Our estimates indicated that the reduction rate of SO<sub>2</sub> emissions was much lower between 2019 and 2022 than that at 53% between 2015 and 2019. In particular, the emissions from the power sector were estimated to decline only 7% during 2019-2022. The result confirmed that the abatement of SO<sub>2</sub> emissions have been clearly decelerated following the full implementation of ultra-low emission retrofits, suggesting that the potential of further reduction of SO<sub>2</sub> emissions for power sectors has become more limited. More energy structure adjustment instead of end-of-pipe controls is needed for the sector.

In contrast to SO<sub>2</sub>, the emissions of NO<sub>x</sub> and PM<sub>2.5</sub> were estimated to decline faster during 2019-2022 than 2015-2019. Industrial sectors contributed largely to these reductions, with the emission declining 27% and 22% for NO<sub>x</sub> and PM<sub>2.5</sub>, respectively (Figure S2). These reductions reflected expansion of intensified pollution control policies from power to other sectors, particularly the ultra-low emission standards implemented for steel (2019) and cement industries (2020) (<https://sthjt.jiangsu.gov.cn/>, last visited on October 2025). By 2022, Jiangsu province had implemented ultra-low emission retrofits in over 80% of iron & steel enterprises and approximately 60% of cement clinker production lines (DEE, 2023). However, slower progress of emission controls in coking, glass, and chemical industries highlighted substantial emission reduction potential in these non-electrical industrial sectors. Meanwhile, the NO<sub>x</sub> emissions of transportation were estimated to decline by



419 41% from 2019 levels (53% for light-duty gasoline vehicles), driven mainly by the  
420 nationwide implementation of China VI vehicle emission standard and increasing  
421 penetration of renewable energy vehicles.

422 NMVOCs, as critical precursors of both secondary  $PM_{2.5}$  and  $O_3$  formation, exhibited  
423 a slower decline in emissions and have emerged as the priority of emission controls in  
424 Jiangsu (Figure S2). Industrial activities dominate NMVOCs emissions in Jiangsu,  
425 contributing 68% of the provincial total emissions. It resulted from the heavy  
426 dependence of the province on chemical industries. For example, the province  
427 accounted for over 40% of national pesticide active ingredient and dye production.  
428 Notably, more than 60% of small-scale chemical enterprises persisted in utilizing  
429 solvent-based coatings, inks, and adhesives with high-VOCs content (Simayi et al.,  
430 2022; Hu et al., 2024). Furthermore, recent expansions in solvent consumption and  
431 chemical output within large-scale enterprises along the Yangtze River have largely  
432 offset the emission reductions through improvement of manufacturing and pollution  
433 control technologies (Li et al., 2019). Consequently, intensified emission controls  
434 should be urgently required for targeting key industrial sectors and critical regions for  
435 NMVOCs reduction. Agricultural  $NH_3$  emissions in Jiangsu have experienced a  
436 decline of 14% during 2019-2022, primarily attributed to reduced nitrogen fertilizer  
437 usage. However, the absence of effective  $NH_3$  control measures prevented further  
438 substantial reduction of emissions for the sector (Zhou et al., 2023; Zhao et al., 2022).

### 439 **3.1.2 Daily emission variability for air pollutants in 2022**

440 Figure 2 show the daily variability of total and sectoral emissions of various pollutants  
441 ( $SO_2$ ,  $NO_x$ ,  $PM_{2.5}$ , NMVOCs, and  $NH_3$ ) in 2022, respectively (the time series of  
442 emissions ( $NO_x$  as an example) for all the involved source categories are provided in  
443 Supplementary Figure S3). The results revealed distinct seasonal emission patterns of  
444 air pollutants driven by anthropogenic activities and/or meteorological conditions.

445 The emissions of  $SO_2$  and primary  $PM_{2.5}$  followed the seasonal patterns of fossil  
446 energy consumption (Yun et al., 2021), with clear peaks in winter (from December to





February) associated with the substantial coal combustion for residential heating and elevated industrial energy demand (Geng et al., 2021; Zhan et al., 2023). Regarding NO<sub>x</sub>, transportation has become the primary contributor to the emissions along with improved emission controls from the power and industrial sectors. Following the lifting of COVID-19 lockdown since June 2022, moreover, residents exhibited a strong desire to travel, which enhanced the emissions from transportation. Compared to the spring (from March to May), NO<sub>x</sub> emissions from transportation increased 12% during the summer (from June to August), consistent with the elevated population mobility (Supplementary Figure S4). Additionally, the NO<sub>x</sub> emission peak in March reflected the resumption of industrial production and construction activities after the Chinese New Year. The area of construction for residential and commercial buildings increased 56% from February to March, with these activities heavily dependent on diesel-powered machinery (Yang et al., 2015; Cliff et al., 2023). The NMVOCs emissions were the largest in summer. Enhanced volatilization of solvents and industrial chemicals by the warmer temperatures resulted in a 22% growth of summer emissions compared to spring. Similar to NO<sub>x</sub>, the NMVOCs emissions in March rebounded with a 17% growth compared to February, reflecting the resumption of coating, printing, and petrochemical industries. NH<sub>3</sub> were closely associated with farming cycles, peaking during Spring sowing and Autumn harvesting periods.

Notably, the province has made great efforts on reducing emissions during the period with heavy pollution weather (DEE, 2022). Compared to August 2022, mandatory restrictions on coal-fired boilers and industrial plants for September resulted in an 11% reduction of coal consumption for major industrial sectors, leading to a decline of 7%, 10%, 15%, and 12% for anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, and NMVOCs, respectively. This demonstrated the effectiveness of pollution control measures conducted by the government on counteracting pollution episodes around August and September, despite persistent meteorological challenges (Wang et al., 2023). However, subsequent emission rebounds in winter for SO<sub>2</sub> (+24% compared with those in Autumn) and PM<sub>2.5</sub> emissions (+19%) underscored the limitation of seasonal control



476 strategies for combustion-derived pollutants, emphasizing the imperative for clean  
477 energy promotion to achieve sustainable emission abatement.

478 In April 2022, a great reduction in air pollutant emissions was estimated. Compared  
479 with March, the emissions of SO<sub>2</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, and NMVOCs decreased by 11%, 8%,  
480 6%, and 12% respectively. This abrupt decline was temporally associated with the  
481 COVID-19 induced lockdown implemented in Shanghai (March 28-June 1, 2022).  
482 The lockdown substantially disrupted industrial production, transportation activities,  
483 and daily routines in neighboring Jiangsu Province. The results showed that  
484 short-term public health incidents exerted profound impact on air pollutant emissions  
485 (Zhang et al., 2024; Ma et al., 2023).

### 486 **3.1.3 High-resolution maps of air pollutant emissions**

487 Based on the real-time geospatial information from the POI system (e.g., quarterly  
488 updated road networks, land use types, and monthly revised construction sites), we  
489 achieved the evolving spatial pattern of daily air pollutant emissions with a horizontal  
490 resolution of 3 km×3 km. Figure 3 presents the spatial distribution of daily average  
491 emissions of major sectors in Jiangsu Province for 2022. We selected NO<sub>x</sub> as an  
492 example to illustrate the sector heterogeneity. The NO<sub>x</sub> emissions from power,  
493 industrial, vehicle, off-road transportation and residential sources in Jiangsu were  
494 calculated at 144, 109, 247, 183 and 45 Gg respectively. Aviation emissions (less than  
495 1% of total NO<sub>x</sub>) were excluded due to their tiny contribution to the total emissions.

496 The spatial pattern of emissions was closely associated with corresponding  
497 anthropogenic activities. Agricultural machinery emissions were predominantly  
498 located in northern agricultural zones and coastal areas, correlating with the  
499 spatiotemporal distribution of farming activities. In contrast, emissions from other  
500 sources were more concentrated in the southern cities, especially along the Yangtze  
501 River with the most abundant power and industrial plants. The NO<sub>x</sub> emissions from  
502 five cities in southern Jiangsu (Nanjing, Suzhou, Wuxi, Changzhou, Zhenjiang)  
503 accounted for 59% and 63% of provincial power and industrial emissions,



504 respectively. On-road transportation emissions demonstrated a strong dependence on  
505 the road network. Nanjing and Xuzhou, as critical national railway transportation hubs,  
506 contributed 24% and 13% of provincial NO<sub>x</sub> emissions from railways (Wang et al.,  
507 2016). In addition, Suzhou contributed 29% of provincial marine emissions, attributed  
508 to its pivotal role in Yangtze River Delta inland waterway logistics (Shen et al., 2021).  
509 Unsurprisingly, the residential NO<sub>x</sub> emissions were closely correlated with the  
510 population density.

#### 511 **3.1.4 Assessment of monthly variability**

512 Figure 4 compares the monthly distributions of SO<sub>2</sub>, NO<sub>x</sub>, and PM<sub>2.5</sub> emissions  
513 estimated in this study with those in MEIC, as well as those of provincial averages of  
514 ambient concentrations of corresponding species obtained from the state-operating  
515 observation sites in Jiangsu. Due to the unavailability of MEIC for the year 2022, we  
516 used the result for 2020 instead.

517 For SO<sub>2</sub> (Figure 4a) and PM<sub>2.5</sub> (Figure 4e), our analysis demonstrated a close  
518 agreement between monthly variation in emissions and that in observed concentration  
519 across Jiangsu Province. The near-real-time emission estimates effectively captured  
520 the short-term fluctuations, including the abrupt reduction in April associated with  
521 COVID-19 lockdown and the seasonal change from the temporary pollution control  
522 measures in autumn. These results partly justified the capability of the approach to  
523 track the effect of changing anthropogenic activities on air pollutant emissions.  
524 Meanwhile, we found contrary monthly distributions between NO<sub>x</sub> emissions and the  
525 observed concentration of NO<sub>2</sub> (Figure 4c). The largest emissions were estimated in  
526 summer months but the lowest concentrations were observed for the same months  
527 across the year. This inconsistency likely resulted from following factors. Increased  
528 transportation activity during summer, particularly mobility rebound after lockdown,  
529 elevated NO<sub>x</sub> emissions, while NO<sub>2</sub> was substantially consumed for O<sub>3</sub> formation  
530 through photochemical reactions. In winter, there was more NO<sub>2</sub> accumulation in the  
531 atmosphere with slower photochemical reactions and reduced boundary layer heights



532 (Ding et al., 2015; Wang et al., 2012). In addition, we found a similar correlation  
533 between  $\text{PM}_{2.5}$ - $\text{NO}_2$  in monthly trends, implying the importance of controlling  $\text{NO}_x$   
534 emissions in reducing  $\text{PM}_{2.5}$  pollution.

535 Similar monthly distribution of emissions were found for the national (MEIC) and  
536 provincial emission estimates (this work), implying regular patterns of monthly  
537 anthropogenic activities could be captured by both inventories. Nevertheless,  
538 disparities existed in the overall emission totals and sector distributions between the  
539 two inventories. For instance, the contributions of industry to provincial emissions of  
540  $\text{SO}_2$  and  $\text{NO}_x$  were estimated at 45% and 15% in this work, greatly different from the  
541 MEIC estimation at 72% and 41%, respectively. These discrepancies might be  
542 attributed to that the national inventory (MEIC) for 2020 has not yet fully included the  
543 information of emission control technology upgrades (e.g., ultra-low emission  
544 retrofits) in the industrial sector. Taking the sintering process in the steel industry as  
545 an example, our facility-level estimations indicated that the average emission factors  
546 for  $\text{SO}_2$ ,  $\text{NO}_x$ , and  $\text{PM}_{2.5}$  were 0.143 kg/t, 0.228 kg/t, and 0.037 kg/t, respectively,  
547 much lower than the recommended values of 1.34 kg/t, 0.55 kg/t, and 2.52 kg/t from  
548 the guidelines for development of national emission inventory (He et al., 2018).

549 Substantial discrepancies were revealed for off-road transportation of  $\text{SO}_2$  emissions.  
550 The provincial  $\text{SO}_2$  emission estimate from marine (12,877 metric tons) were almost  
551 three times of that by MEIC (4,690 metric tons). As a major freight hub in the eastern  
552 coastal region of the country, Jiangsu Province played a pivotal role in marine  
553 transportation, and approximately 60% of vessels utilized heavy oil with high-sulfur  
554 content as fuel (Dong et al., 2025). Application of national average EFs for the sector  
555 might lead to underestimation in emissions. Furthermore, the national inventory  
556 ignored the emissions from passing vessels at ports. Inclusion of such vessels would  
557 increase the  $\text{SO}_2$  emissions in the Yangtze River Delta region by a factor of 2.3  
558 (Zhang et al., 2017). As power and industrial sectors have gradually completed  
559 ultra-low emission retrofits, marine emissions with less stringent controls may  
560 become more important in the future, requiring greater efforts on fuel quality



561 improvement and stricter emission controls.

### 562 **3.2 Impacts of short-term lockdown on changes in emissions**

563 From March 28 to June 1 in 2022, Shanghai, the largest megacity in YRD and the  
 564 national center of economy, finance, manufacturing, and maritime trade in China,  
 565 implemented stringent COVID-19 lockdown measures that suspended intercity  
 566 mobility and industrial production and kept only essential logistics. This  
 567 unprecedented lockdown not only disrupted social and economic activities of  
 568 Shanghai, but also brought substantial effects for neighboring regions. Jiangsu  
 569 Province, a highly industrialized region adjacent to Shanghai, experienced severe  
 570 disruptions across service sectors, manufacturing supply chains, and maritime  
 571 logistics, resulting in substantial declines in energy consumption, industrial output,  
 572 and transportation activities. To further quantify the lockdown effect on air pollutant  
 573 emissions, we conducted a comparative analysis between two periods: the  
 574 lockdown-affected period (April-May 2022) and the post-pandemic period, the same  
 575 months one year later (April-May 2023).

576 The first column of Figure 5 (a1, b1, c1, d1) illustrates the variability in daily  
 577 emissions of NO<sub>x</sub>, SO<sub>2</sub>, PM<sub>2.5</sub>, and NMVOCs in Jiangsu during April-May 2022  
 578 (lockdown period) versus 2023 (recovery period), as well as the difference between  
 579 the two periods. The emission differences (calculated as the relative change compared  
 580 to the 2023 level) reached 8%, 6%, 6%, and 10% for these air pollutants, respectively.  
 581 The most substantial decline in pollutant emissions occurred in April 2022, with a  
 582 gradually diminishing difference in May. However, the emissions by the end of May  
 583 2022 did not reach the level of recovery period in May 2023, reflecting the effect of  
 584 temporary measures on reducing economic activities even after the lifting of the  
 585 lockdown. The full economy recovery was delayed until 2023 when pandemic  
 586 restrictions were completely lifted (Li et al., 2023).

587 The second and third columns of Figure 5 (a2-d2 and a3-d3) illustrate the  
 588 contributions of various pollution source categories to the differences in emissions



589 between April-May of 2022 and 2023. Agricultural production remained basically  
590 unaffected by the pandemic, thus the emission changes from agricultural machinery  
591 were not included. The total reduction in NO<sub>x</sub> emissions was 9,970 metric tons,  
592 predominantly attributed to transportation sources. The sector contributed to over 70%  
593 of the emission reduction, including on-road transportation (15%), construction  
594 machinery (27%), marine (19%), railway (5%), and aviation (4%). This result is  
595 consistent with the findings on the effect of the 2020 COVID-19 lockdown (Lv et al.,  
596 2020; Zhao et al., 2020a). However, there was a slight rebound in motor vehicle  
597 emissions in May, which could be associated with basic everyday living and working  
598 needs. Notably, construction machinery and marine were more affected by the  
599 lockdown, attributable to construction material shortages (39% fewer of constructing  
600 and building activities) and disrupted inland waterway logistics (20% less of port  
601 throughput). Compared with transportation, the reduction of NO<sub>x</sub> emissions from the  
602 power (1,955 metric tons) and the industrial sector (1,202 metric tons) were smaller.  
603 The decline in industrial electricity demand reduced the fossil fuel consumption and  
604 thereby the NO<sub>x</sub> emissions from the power sector. During industrial shutdowns and  
605 production restrictions caused by the epidemic, frequent start-ups and shutdowns of  
606 production and pollution control equipment resulted in a clear decline in NO<sub>x</sub>  
607 removal efficiency compared with normal operation condition of selective catalytic  
608 reduction (SCR) systems. Previous measurements found that the average NO<sub>x</sub>  
609 removal efficiency of coal-fired units in iron & steel production enterprises decreased  
610 from 78% to 61% (Shao et al., 2023), which to some extent offset the emission  
611 reduction effect of industrial sources due to production restrictions.

612 SO<sub>2</sub> emission reductions predominantly originated from power (521 metric tons, 21%)  
613 and industrial sectors (1,710 metric tons, 68%). For PM<sub>2.5</sub>, transportation contributed  
614 56% to the total reduction of 3,583 metric tons, with the contributions from on-road  
615 transportation, construction machinery, marine, railway, and aviation accounting for  
616 8%, 18%, 14%, 9%, and 7%, respectively. The emission reductions of NMVOCs were  
617 estimated at 20,170 metric tons. The contribution of industrial sources reached 93%,



618 largely due to a 64% decline in crude oil processing in Jiangsu Province compared to  
 619 2023, as well as the substantial declines in the production of chemical products (e.g.,  
 620 27% less in chemicals fibers and 65% less in ethylene manufacturing, NBS, 2023).  
 621 The results emphasized the lockdown impact on petrochemical industries reliant on  
 622 cross-regional material flows. In contrast, the emissions from residential sector were  
 623 larger for the lockdown period, with its coal consumption 7% more than that in  
 624 recovery period one year later, likely driven by the enhanced heating/cooking  
 625 demands during mobility restrictions.

626 In a summary, the results revealed complicated and diverse interventions of public  
 627 health incidents on energy use and activities for different sectors. The near-real-time  
 628 techniques developed in this work proved capable to capture the fast response of air  
 629 pollutant emissions to the short-term measures conducted during unexpected incidents,  
 630 and to clear identify the driving sectors of emission changes compared to the normal  
 631 conditions.

### 632 **3.3 Evaluation of the near-real-time emission estimates with air** 633 **quality simulation**

634 The near-real-time estimates of provincial emissions were evaluated with air quality  
 635 simulation with CMAQ. To assess model performance, the observed concentrations of  
 636 hourly SO<sub>2</sub>, NO<sub>2</sub>, PM<sub>2.5</sub>, and MDA8 O<sub>3</sub> were compared with the simulations based on  
 637 the provincial-level near-real-time emission estimates and MEIC for the selected four  
 638 months of 2022, as summarized in Supplementary Table S2. Overall, the simulation  
 639 with the provincial emission estimates shows acceptable agreement with the  
 640 observations, with the annual means of NMB and NME ranging -37.1% – 24.1% and  
 641 33.7% – 53.5% for SO<sub>2</sub>, -20.2% – 27.0% and 15.9% – 36.2% for NO<sub>2</sub>, -18.6% – 10.8%  
 642 and 37.5% – 62.5% for PM<sub>2.5</sub>, and -41.2% – -23.1% and 32.7% – 49.3% for O<sub>3</sub>. The  
 643 analogous numbers for MEIC were -33.4% – 25.5% and 40.9% – 51.8% for SO<sub>2</sub>, -19.9%  
 644 – 35.6% and 22.3% – 55.1% for NO<sub>2</sub>, -8.6% – 25.2% and 37.5% – 52.5% for PM<sub>2.5</sub>,  
 645 and -39.9% – -28.1% and 44.3% – 54.5% for O<sub>3</sub>, respectively. Most of the NMB and



646 NME were within the recommended criteria ( $-30\% \leq \text{NMB} \leq 30\%$  and  $\text{NME} \leq 50\%$ ,  
647 Emery et al., 2017). Better performance was achieved using the provincial emission  
648 estimates developed in this work, implying the benefit of applying the refined  
649 emission data on high-resolution air quality simulation.

650 Figures 6 and 7 compares the simulated daily  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentrations based on  
651 the provincial (this work) and national emission estimates (MEIC) against  
652 observations (results for  $\text{SO}_2$  and  $\text{NO}_2$  are shown in Supplementary Figures S5 and S6,  
653 while spatial distributions for all the four pollutants are provided in Supplementary  
654 Figures S7-S10). Compared to MEIC, the provincial-scale emission estimates  
655 demonstrated better model performance in capturing the daily variability of pollutant  
656 concentrations. The greater correlation coefficients (R) between simulated and  
657 observed concentrations based on the near-real-time estimates indicated a remarkable  
658 improvement for all the involved air pollutants (Table S2).

659 For  $\text{PM}_{2.5}$ , the improvement in model performance based on the provincial emission  
660 estimates was particularly prominent concerning the impact of the COVID-19  
661 lockdown measures in April. As shown in Figure 6, the near-real-time approach more  
662 accurately captured the decline in  $\text{PM}_{2.5}$  level from reduced emissions. As a  
663 comparison, notable overestimation of  $\text{PM}_{2.5}$  concentration occurred for simulation  
664 with MEIC. As a national emission inventory, MEIC commonly applied the temporal  
665 profiles of activity data for various sectors for the whole country, thus could  
666 insufficiently track the effect of temporary and unexpected events on emissions, such  
667 as the city lockdown. For  $\text{O}_3$ , despite of the underestimation for both emission  
668 inventories, application of the near-real-time provincial estimates not only reduced the  
669 underestimation compared to MEIC but also better captured the variability of  $\text{O}_3$   
670 concentration driven by short-term emission fluctuations. These results collectively  
671 demonstrated the improvement in model performance and advantage of near-real-time  
672 emission estimates to support high-resolution air quality simulation.

673





### 674 **3.4 Impact of daily emission change on the variability of PM<sub>2.5</sub> and O<sub>3</sub>** 675 **concentrations**

#### 676 **3.4.1 Anthropogenic-driven contributions to variability of PM<sub>2.5</sub> and MDA8 O<sub>3</sub>** 677 **concentrations**

678 Figure 8 presents the contributions of the changing daily emissions to the monthly  
 679 variability of PM<sub>2.5</sub> and MDA8 O<sub>3</sub> concentrations based on the MLR model. The  
 680 model performance was assessed with observed PM<sub>2.5</sub> and O<sub>3</sub> concentrations  
 681 (Supplementary Figure S11). The simulated concentrations were strongly correlated  
 682 with observational data, with the correlation coefficient (R) of 0.79 for PM<sub>2.5</sub> and 0.88  
 683 for MDA8 O<sub>3</sub>. The validation indicated satisfying performance of MLR in capturing  
 684 provincial air quality variability.

685 The anthropogenic-driven variability of PM<sub>2.5</sub> concentration was basically consistent  
 686 with the temporal variation of estimated emissions. As shown in Figure 8a, the  
 687 abundant emissions in January resulted in a prominent enhancement of 12.7 µg/m<sup>3</sup> for  
 688 PM<sub>2.5</sub> concentration, followed by December (1.8 µg/m<sup>3</sup>) and June (1.6 µg/m<sup>3</sup>). In  
 689 particular, the enhancement of June was driven largely by the post-pandemic  
 690 economic recovery, as discussed in in Section 3.2. For most warm months (April to  
 691 October, except June), negative impacts of anthropogenic activities on PM<sub>2.5</sub> level  
 692 were found, ranging 1.1 – 4.2 µg/m<sup>3</sup>. Clear decline of PM<sub>2.5</sub> due to emission change  
 693 was also found in February (5.5 µg/m<sup>3</sup>), resulting probably from the greatly reduced  
 694 human activities (industry and transportation) during the Chinese New Year holiday.  
 695 The PM<sub>2.5</sub> growth occurred during winter heating period highlighted the necessity of  
 696 accelerating transition of clean household energy and improving management of  
 697 industrial production after the short-term lockdowns.

698 The variation of anthropogenic emissions was found to elevate O<sub>3</sub> concentrations in  
 699 most months of the year, particularly for warm seasons (Figure 8b). The  
 700 enhancements during March-August ranged 0.8 – 3.8 µg/m<sup>3</sup>, suggesting the important  
 701 role of human activities in aggravating O<sub>3</sub> pollution. High temperature in summer



702 promoted the emissions of temperature-dependent O<sub>3</sub> precursors, particularly  
703 NMVOCs from various sources (Figure 2d). In addition, the NO<sub>x</sub> emissions from  
704 certain were elevated in warm seasons, e.g., those from off-road machinery in the  
705 summer harvest season (Figure 2a). The growing abundance of precursors, together  
706 with high temperature, enhanced the photochemical production rate of O<sub>3</sub>.

707 However, the anthropogenic emissions during winter demonstrated a net negative  
708 contribution to surface O<sub>3</sub> concentrations (e.g., -6.2 and -2.4 µg/m<sup>3</sup> for November and  
709 December, respectively), indicating a shift in the chemical regime of O<sub>3</sub> formation.  
710 This phenomenon primarily resulted from enhanced NO<sub>x</sub> titration amplified by  
711 elevated NO<sub>x</sub> level. Simultaneously, reduced NMVOCs emissions and diminished  
712 photochemical activity restricted the efficiency of radical-driven O<sub>3</sub> production. The  
713 resulting O<sub>3</sub>-depleting reactions overwhelmed potential formation mechanisms,  
714 leading to the estimated negative contribution from anthropogenic emissions. This  
715 pattern contrasted sharply with the net positive effect of anthropogenic activities in  
716 summer months, and underscored the complex season-dependent response of O<sub>3</sub> level  
717 to the changing precursor emissions.

#### 718 **3.4.2 Impact of fluctuations in anthropogenic emissions by precursor and sector** 719 **on PM<sub>2.5</sub> and MDA8 O<sub>3</sub> concentrations**

720 The impacts of anthropogenic emission fluctuations on variability of PM<sub>2.5</sub> and O<sub>3</sub>  
721 concentrations were quantified by precursor and sector, with a machine learning  
722 framework integrating XGBoost and SHAP analysis. Derived from the 10-fold cross  
723 validation, the correlation coefficient (R) between machine learning prediction and  
724 observation reached 0.78 and 0.81 for daily PM<sub>2.5</sub> and MDA8 O<sub>3</sub>, respectively,  
725 suggested satisfying capability of the machine learning framework in predicting the  
726 anthropogenic-driven variability of PM<sub>2.5</sub> and O<sub>3</sub> concentrations (Supplementary  
727 Figure S12).

728 Figure 9a and 9b illustrates the contributions of changing emissions from different  
729 pollutant-sector combinations to the variability of PM<sub>2.5</sub> concentration in January and



730 that of MDA8 O<sub>3</sub> in July, respectively. The temporal variability of PM<sub>2.5</sub> level  
731 attributable to anthropogenic emission changes was in general consistent with that of  
732 observed surface PM<sub>2.5</sub> concentration (Figure 9a). For O<sub>3</sub>, there existed some  
733 discrepancy between the temporal distribution of anthropogenic-driven variability and  
734 observed concentration in summer. This discrepancy may be attributed to the  
735 substantial impacts of meteorological conditions and biogenic VOCs emissions on O<sub>3</sub>  
736 formation (Gu et al., 2023).

737 Among all the pollutant-sector combinations, fluctuations in agricultural NH<sub>3</sub>  
738 emissions accounted for 67.3% of the variability of PM<sub>2.5</sub> concentrations in January,  
739 followed by off-road NO<sub>x</sub> (12.9%) and residential PM<sub>2.5</sub> emissions (4.9%). The  
740 contribution of NH<sub>3</sub> emission variation significantly exceeded those of NO<sub>x</sub> (17.7%),  
741 PM<sub>2.5</sub> (10.8%), and SO<sub>2</sub> (4.2%), suggesting that Jiangsu may be transitioning to an  
742 NH<sub>3</sub>-rich regime following substantial reductions in SO<sub>2</sub> and NO<sub>x</sub> emissions (Zhao et  
743 al., 2020b). Therefore, agricultural NH<sub>3</sub> control has become the priority of the strategy  
744 design for PM<sub>2.5</sub> pollution alleviations, compared to traditional NO<sub>x</sub> abatement. The  
745 fluctuations in VOC-Industry contributed to 48.5% of the variability of MDA8 O<sub>3</sub>  
746 concentrations in July, followed by off-road VOCs (9.7%) and NO<sub>x</sub> emissions (8.9%).  
747 In total, the NMVOCs accounted for 69.7% of the anthropogenic-driven variability of  
748 O<sub>3</sub> concentration, exceeding the contributions from NO<sub>x</sub> (14.5%), PM<sub>2.5</sub> (11.0%), and  
749 SO<sub>2</sub> (4.9%). The positive contribution of NO<sub>x</sub> to MDA8 O<sub>3</sub> indicated that the O<sub>3</sub>  
750 formation mechanism in Jiangsu may be shifting from a VOCs-limited regime  
751 towards a transitional or NO<sub>x</sub>-limited regime. Regarding the sector contributions with  
752 various species aggregated, the agricultural emission fluctuations contributed most to  
753 anthropogenic-driven variability of PM<sub>2.5</sub> concentration (67.3%, Figure 9c), while  
754 industrial activities contributed most to that of O<sub>3</sub> concentration (54.8%, Figure 9d).  
755 Notably, off-road transportation emerged as an important contributor to both  
756 pollutants (15.6% for PM<sub>2.5</sub> and 24.4% for O<sub>3</sub>), providing clear evidence for policy  
757 making of coordinating control of PM<sub>2.5</sub> and O<sub>3</sub> pollution.



#### 758 4. Conclusion remarks

759 In this study, we incorporated near-real-time activity data from multiple sources and  
760 developed a method for continuously estimating the regional daily air pollutant  
761 emissions of anthropogenic origin. We then applied this method to estimate the  
762 spatiotemporal evolution of emissions in Jiangsu Province, a typical developed area in  
763 eastern China, with a particular focus on the period during the COVID-19 lockdown  
764 in 2022 and the corresponding phase after the lifting of restrictions in 2023. Finally,  
765 we constructed a rapid assessment approach that utilized machine learning algorithms  
766 to quantify the impact of fast changing emissions on variability of daily air quality.  
767 Our research indicated that emission controls have played a crucial role in abatement  
768 of air pollutant emissions. The provincial emissions of SO<sub>2</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, NMVOCs,  
769 and NH<sub>3</sub> decreased 17%, 33%, 18%, 7%, and 11%, respectively, from 2019 to 2022.  
770 Implementation of ultra-low emission retrofits for industrial sectors has proven  
771 effective in reducing primary PM<sub>2.5</sub> and NO<sub>x</sub> emissions. However, there is an urgent  
772 need to enhance NMVOCs emission control in key industrial sectors and areas. We  
773 identified distinct temporal variabilities of emissions for various air pollutants. The  
774 emissions of SO<sub>2</sub> and PM<sub>2.5</sub> were influenced greatly by fossil fuel consumption  
775 pattern, while NO<sub>x</sub> emissions were increasingly dominated by that of transportation.  
776 The NMVOCs emissions peaked in the summer and declined in winter, followed by a  
777 rebound in emissions after the Chinese New Year. Our comparative analysis indicated  
778 that the emissions of NO<sub>x</sub>, SO<sub>2</sub>, PM<sub>2.5</sub>, and NMVOCs in Jiangsu during the  
779 COVID-19 lockdown of Shanghai in April-May 2022 were respectively 8%, 6%, 6%,  
780 and 10% smaller than those in the same months of 2023. Transportation was identified  
781 as the primary contributors to the reductions in NO<sub>x</sub> and PM<sub>2.5</sub> emissions, while  
782 industry accounted for 93% of the reduction in NMVOCs, closely associated with the  
783 disrupted cross-regional product supply chains. Indicated by the contributions of  
784 changing emissions from pollutant-sector combinations to the variability of PM<sub>2.5</sub> and  
785 O<sub>3</sub> levels, reducing agricultural NH<sub>3</sub> emissions should be critical for PM<sub>2.5</sub> pollution  
786 alleviation, and off-road transportation has become a priority target for coordinating



787 control of both PM<sub>2.5</sub> and O<sub>3</sub> pollution. The outcomes demonstrated the importance of  
788 near-real-time techniques on tracking the fast-changing air pollutant emissions,  
789 identifying the driving factors of air pollution variability, and supporting the policy  
790 making of air quality management.

791 The limitations of this work existed mainly in the near-real-time information of  
792 multiple sources and the rapid assessment of air quality variability. For instance,  
793 CEMS covered only relatively big point sources, thus we had to assume that the small  
794 and fugitive sources followed similar variability of emissions with point sources. As  
795 CEMS only covers SO<sub>2</sub>, PM<sub>2.5</sub>, and NO<sub>x</sub>, the use of electricity consumption data for  
796 NMVOCs may introduce substantial uncertainty. Future improvement in online  
797 monitoring of NMVOCs will enhance the estimation of temporal variation of  
798 emissions. Moreover, the machine learning process ignored the contributions from  
799 regional transport, which could result in some bias in analyzing the impacts of  
800 anthropogenic emissions on air quality. However, in contrast to time-consuming  
801 numerical modeling, machine learning offered a rapid and reliable assessment of the  
802 impact of daily emission changes on air quality, which exactly addressed the  
803 requirement of air quality management, and was recommended in future policy  
804 making of air pollution controls.

## 805 **Data availability**

806 The gridded emission data for Jiangsu Province 2022-2023 can be downloaded at  
807 <http://www.airqualitynju.com>

## 808 **Author contributions**

809 CGu developed the methodology, conducted the research and wrote the draft. YZhao  
810 and LZhang developed the strategy and designed the research, and YZhao revised the  
811 manuscript. YWang provided the support of machine learning modeling. YJi provided  
812 the support of WFR-CMAQ. ZZhang, and WZhao supported emission data processing.  
813 SSun, YBian, JZhu, and SZhong provided the support of emission data.



## 814 **Competing interests**

815 The authors declare that they have no conflict of interest.

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## 1195 **Figure captions**

1196 **Figure 1** The research framework of near-real-time emission estimation and  
 1197 application in this work.

1198 **Figure 2** Daily emission estimates of anthropogenic air pollutants by sector for  
 1199 Jiangsu Province in 2022. (a) NO<sub>x</sub>; (b) SO<sub>2</sub>; (c) PM<sub>2.5</sub>; (d) NMVOCs; (e) NH<sub>3</sub>.

1200 **Figure 3** Spatial distribution of anthropogenic NO<sub>x</sub> emissions for Jiangsu Province in  
 1201 2022 with a horizontal resolution of 3×3 km. (a) Total emissions; (b) Power; (c)  
 1202 Industry; (d) Vehicle; (e) Off-road transportation; (f) Residential. The map data  
 1203 provided by Resource and Environment Data Cloud Platform are freely available for  
 1204 academic use (<http://www.resdc.cn/data.aspx?DATAID=201>), © Institute of  
 1205 Geographic Sciences & Natural Resources Research, Chinese Academy of Sciences.

1206 **Figure 4** The monthly air pollutant emissions for Jiangsu Province in 2022 estimated  
 1207 in this study (a, c, and e) and in national emission inventory (MEIC; b, d, and f). The  
 1208 emissions of SO<sub>2</sub> (a and b), NO<sub>x</sub> (c and d) and primary PM<sub>2.5</sub> (e and f) are contained.  
 1209 The red lines with triangles represent the observed monthly surface concentrations of  
 1210 corresponding air pollutants.

1211 **Figure 5** The differences between the emissions of NO<sub>x</sub> (a), SO<sub>2</sub> (b), PM<sub>2.5</sub> (c) and  
 1212 NMVOCs (d) in April-May for 2022 and 2023 in Jiangsu Province. The first column  
 1213 illustrates the daily total emissions and the differences for the period of the two years.  
 1214 The second column illustrates the contributions of various source categories to the  
 1215 differences in daily total emissions, and the third column aggregates them for the  
 1216 whole period.

1217 **Figure 6** The comparison between the observed daily PM<sub>2.5</sub> concentrations and those  
 1218 simulated with different emission inventories (this work and MEIC) for January, April,  
 1219 July and October 2022 for Jiangsu Province.

1220 **Figure 7** The comparison between the observed daily O<sub>3</sub> concentrations and those  
 1221 simulated with different emission inventories (this work and MEIC) for January, April,



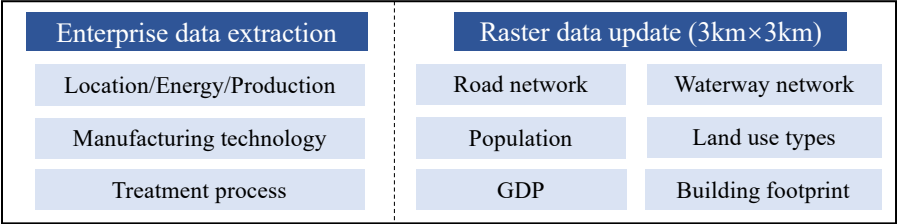
1222 July and October 2022 for Jiangsu Province.

1223 **Figure 8** The monthly anomaly in  $PM_{2.5}$  (a) and MDA8  $O_3$  concentrations (b) driven  
1224 by the changing daily emissions for Jiangsu Province in 2022, based on the MLR  
1225 model.

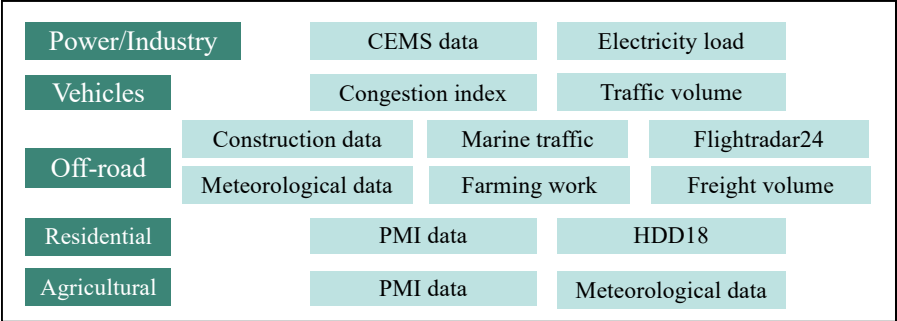
1226 **Figure 9** Anthropogenic pollutant and sector drivers of  $PM_{2.5}$  and MDA8  $O_3$   
1227 variability. (a) and (b) illustrate the contributions of pollutant-sector combinations to  
1228 the variability of  $PM_{2.5}$  in January and that of  $O_3$  in July, derived from SHAP analysis.  
1229 The black dashed lines represent the observed daily ground-level concentrations of  
1230  $PM_{2.5}$  and MDA8  $O_3$ . (c) and (d) provided the contributions of the changing emissions  
1231 from different sectors, with those of various precursor species aggregated.



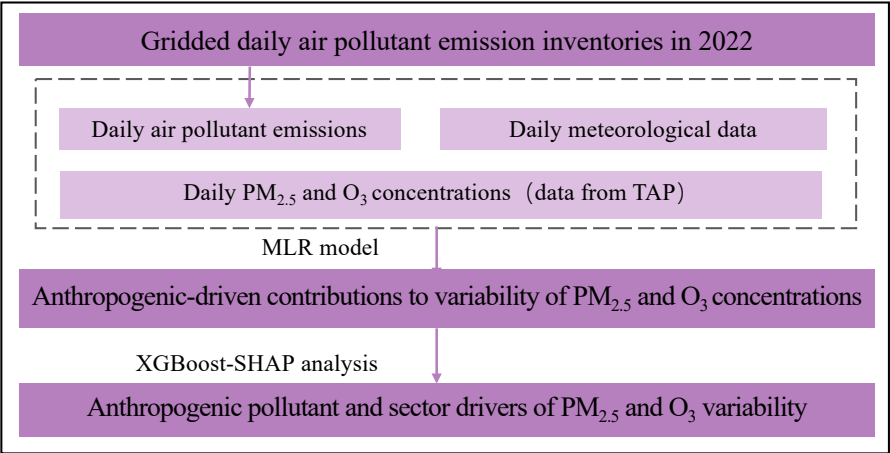
Basic data collection and processing



Near-real-time activity data of multiple sources

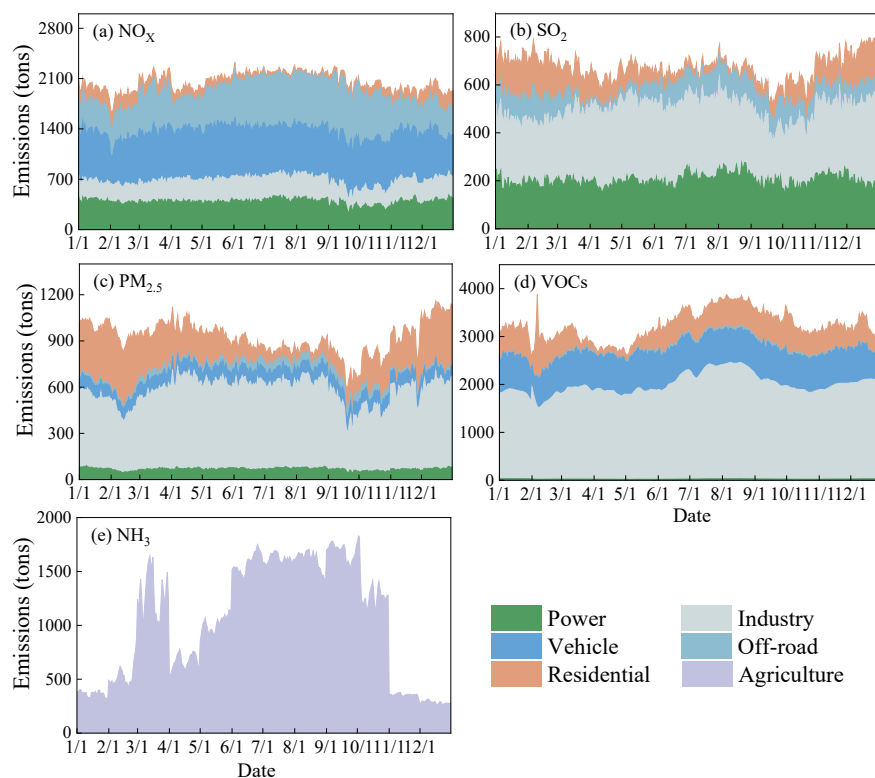


Construction and application of near real-time emissions

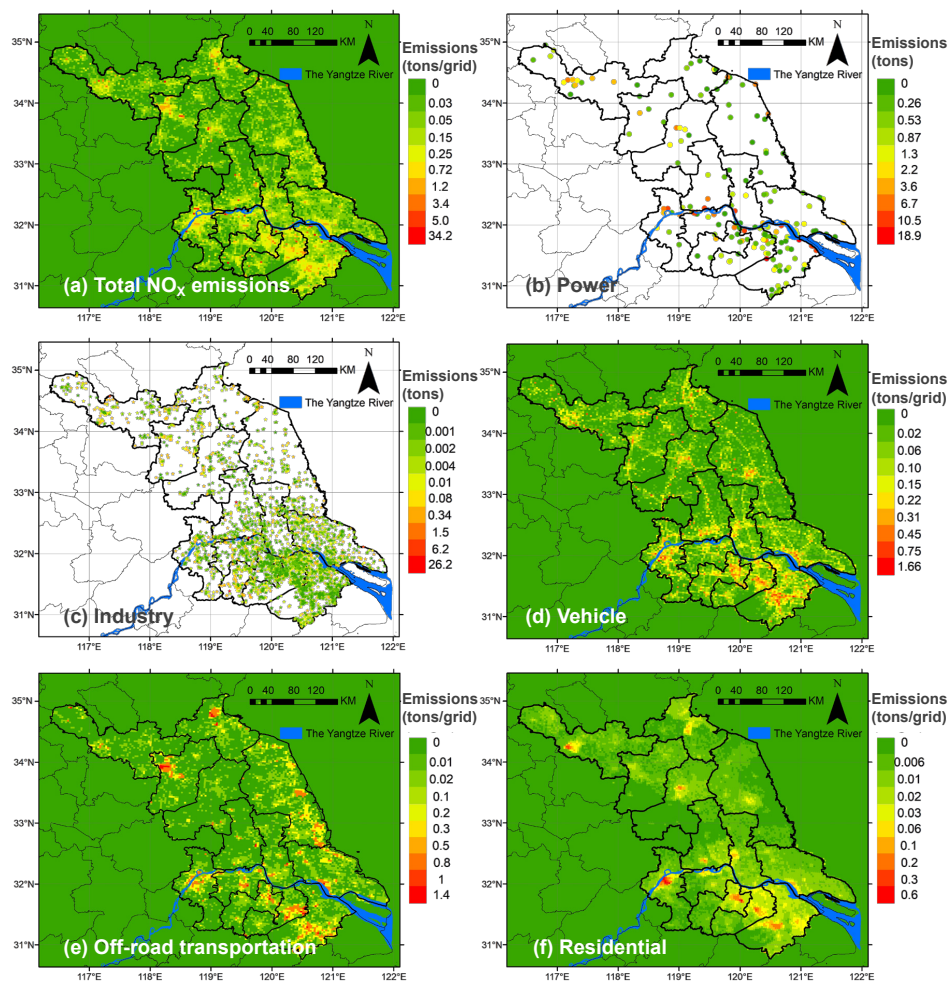


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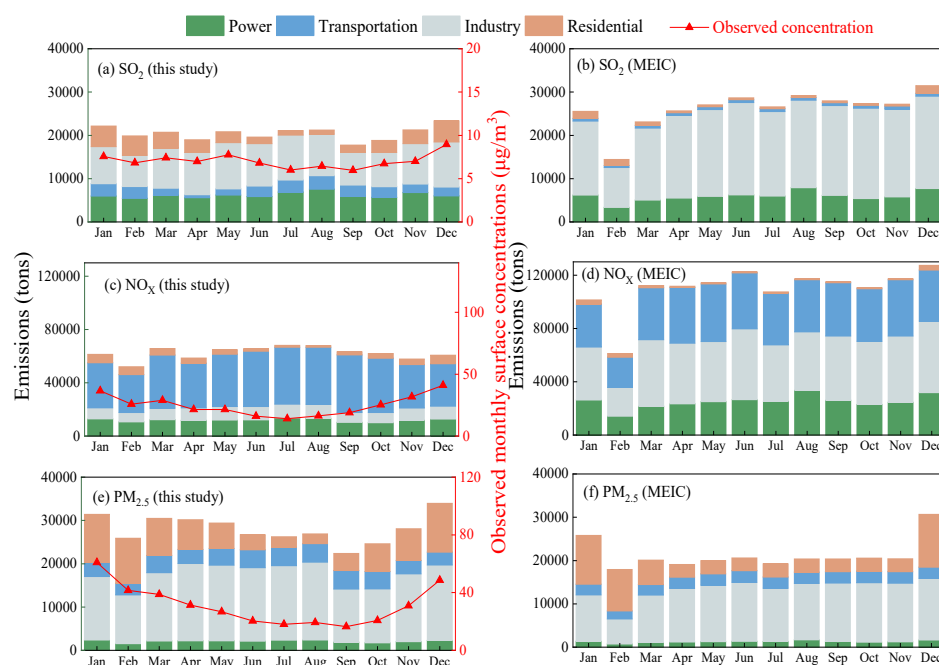
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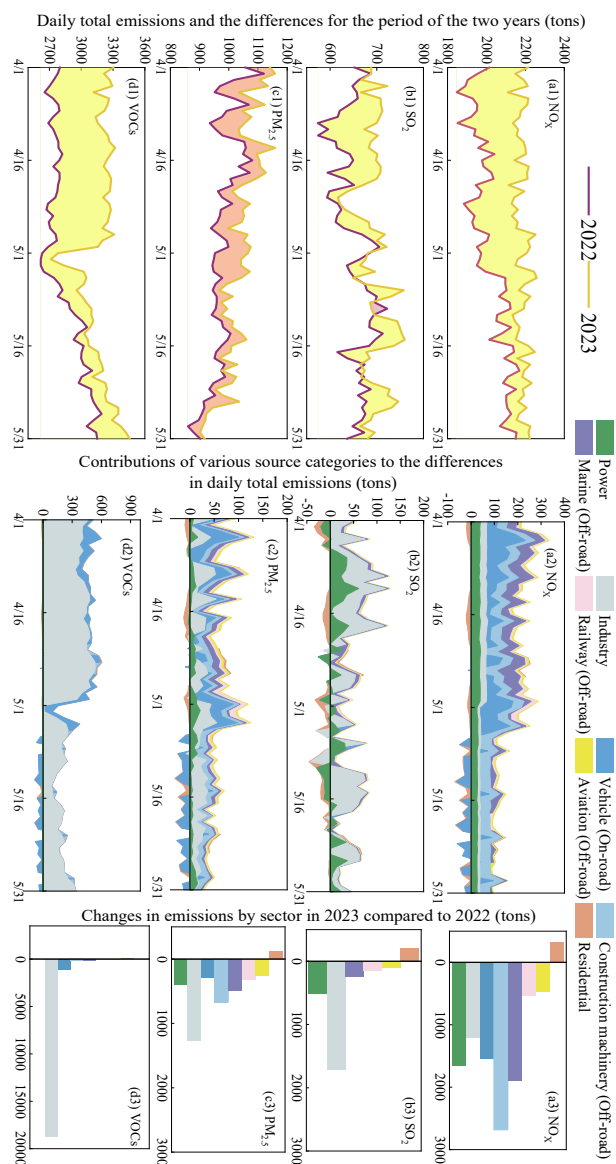


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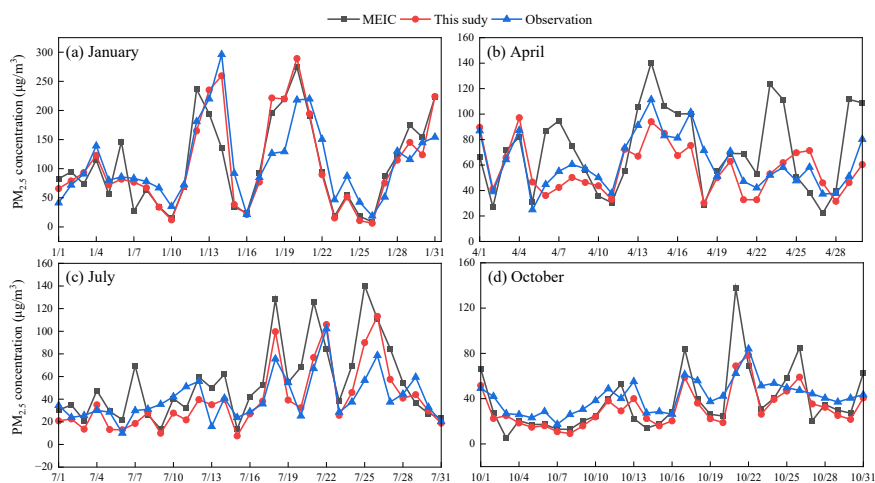


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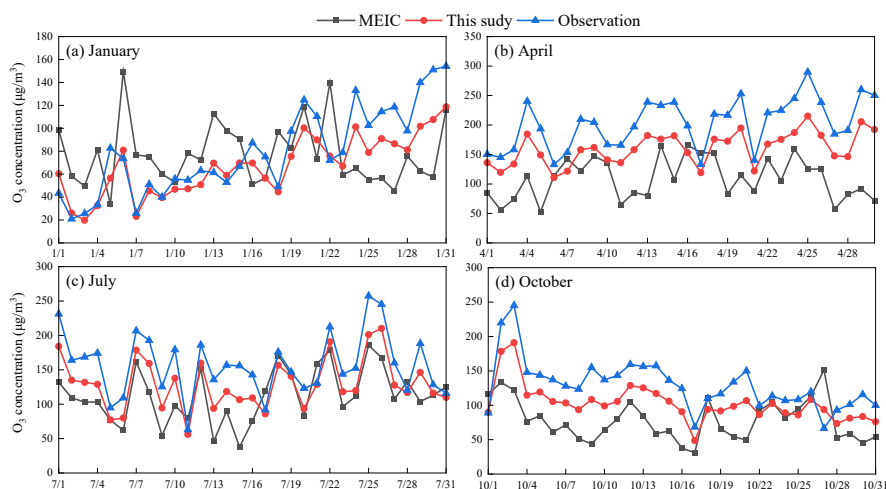




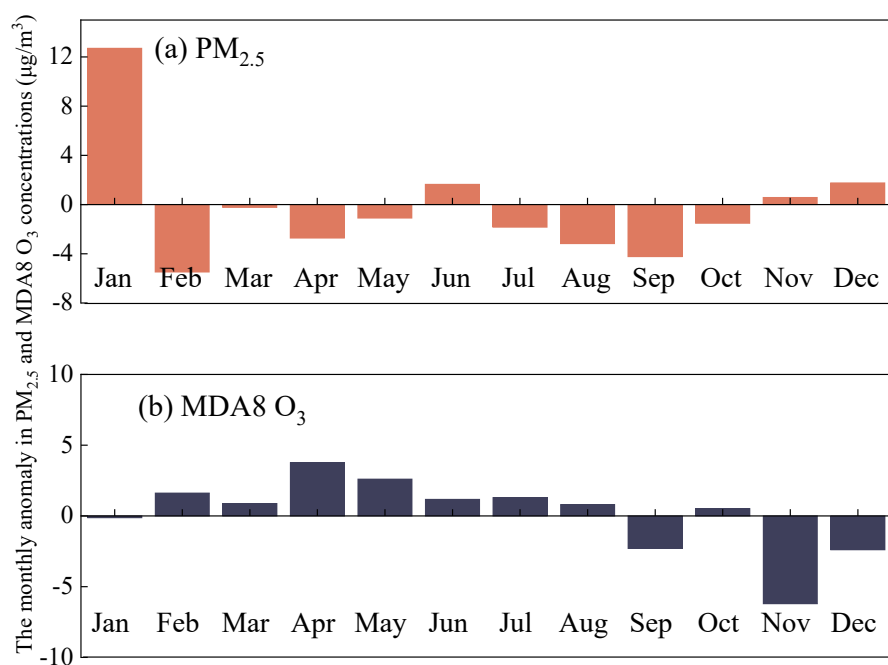
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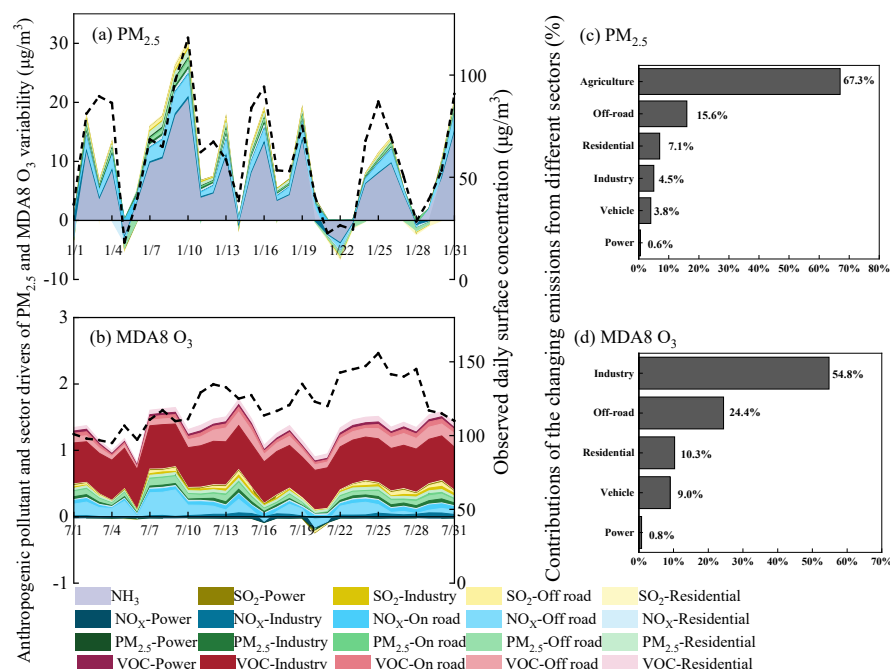
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