Unbalanced emission reductions of different species and sectors in China during COVID-19 lockdown derived by multi-species surface observation assimilation

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Abstract. The unprecedented lockdown of human activities during the COVID-19 pandemic have significantly influenced the social life in China. However, understanding of the impact of this unique event on the emissions of different species is still insufficient, prohibiting the proper assessment of the environmental impacts of COVID-19 restrictions. Here we developed a multi-air pollutant inversion system to simultaneously estimate the emissions of NO_x, SO_2, CO, $PM_{2.5}$ and PM_{10} in China during COVID-19 restrictions with high temporal (daily) and horizontal (15km) resolutions. Subsequently, contributions of emission changes versus meteorology variations during COVID-19 lockdown were separated and quantified. The results demonstrated that the inversion system effectively reproduced the actual emission variations of multi-air pollutants in China during different periods of COVID-19 lockdown, which indicate that the lockdown is largely a nationwide road traffic control measurement with NO_x emissions decreased substantially by ~40%. However, emissions of other air pollutants were found only decreased by ~10%, both because power generation and heavy industrial processes were not halted during lockdown, and residential activities may actually have increased due to the stay-at-home orders. Consequently, although obvious reductions of $PM_{2.5}$ concentrations occurred over North China Plain (NCP) during lockdown period, the emission change only accounted for 8.6% of $PM_{2.5}$ reductions, and even led to substantial increases of O_3. The meteorological variation instead dominated the changes in $PM_{2.5}$ concentrations over NCP, which contributed 90%

37 of the PM$_{2.5}$ reductions over most parts of NCP region. Meanwhile, our results also suggest that the local stagnant

38 meteorological conditions together with inefficient reductions in PM$_{2.5}$ emissions were the main drivers of the unexpected

39 COVID-19 haze in Beijing. These results highlighted that traffic control as a separate pollution control measure has limited

40 effects on the coordinated control of O$_3$ and PM$_{2.5}$ concentrations under current complex air pollution conditions in China.

41 More comprehensive and balanced regulations for multiple precursors from different sectors are required to address O$_3$ and

42 PM$_{2.5}$ pollution in China.

1 Introduction

44   A novel coronavirus disease (COVID-19) broke out in Wuhan at the end of 2019 but quickly spread across the whole

45 China within a month. To curb the spread of the virus, strict epidemic control measures were implemented by Chinese

46 governments to prevent large gatherings, including strict travel restriction, shutting down of non-essential industries,

47 extended holidays, closing of schools and entertainment houses(Cheng et al., 2020). These restrictions have had a significant

48 impact on the industrial activities and social life, as exemplified by the drop of China's industrial output by 15-30%

49 (https://data.stats.gov.cn/, last accessed on 22 Oct, 2022) and the dramatic decrease of traffic flow by 60–90% in major cities

50 of China during COVID-19 epidemic (http://jiaotong.baidu.com/, last accessed on 22 Oct, 2022), which provides us a natural

51 experiment to examine the responses of the emissions and air quality on the changes in human activities.

52   It has been well documented that the short-term stringent emission control targeted on power generator or heavy

53 industry enacted by Chinese government during certain societal events, such as the 2008 Olympics Games, 2014 Asia-

54 Pacific Economic Cooperation conference and 2015 China Victory Day Parade, is an effective way to reduce emissions and

55 improve air quality (Okuda et al., 2011; Wang et al., 2014; Tang et al., 2015; Zhang et al., 2016; Wu et al., 2020; Chu et al.,

56 2018). However, different from those stringent emission controls, the COVID-19 restrictions are inclined to affect emissions

57 from sectors more closely to social life whose influence on emissions has still not well been assessed. Previous studies

58 suggest that the COVID-19 restrictions have substantially reduced the China's anthropogenic emissions from almost all

59 sectors (Zheng et al., 2021; Huang et al., 2021; Xing et al., 2020). For example, by using a bottom-up method based on near-

60 real-time activity data, Zheng et al. (2021) reported that the emissions of NO$_x$, SO$_2$, CO and primary PM$_{2.5}$ decreased by 36%,

61 27%, 28% and 24% during COVID-19 restrictions, mostly due to the reductions in industry and transportation sector. Xing

62 et al. (2020), by using a response model, estimated stronger COVID-19 shutdown effects on emissions over the North China

63 Plain (NCP) with emissions of NO$_x$, SO$_2$ and primary PM$_{2.5}$ dropped by 51%, 28% and 63%, respectively. Others argue that

64 the COVID-19 restriction may mainly affect the emissions from transportation, light industry and manufacturing, while it

65 has much smaller effects on the emissions from the power generator and heavy industry because of their non-interruptible

66 processes (Chu et al., 2021; Hammer et al., 2021; Le et al., 2020; Zhao et al., 2020). Moreover, the residential emissions may

67 even increase during the COVID-19 lockdown due to the increased demanding for space heating and cooking with the stay-

68 at-home orders. Therefore, Le et al. (2020) only considered the NO$_x$ reductions during COVID-19 restrictions in their

69   investigation of the severe haze during COVID-19 lockdown, and similarly, Hammer et al. (2021) only considered the

70   emission reductions in the transportation sector. This indicates that there has large uncertainty in the current understanding of

71   the effects of COVID-19 restrictions on the emissions of different species.

72   Quantification of the emission changes of different species and different sectors during the COVID-19 lockdown is thus

73   necessary for the comprehensive understanding of the environmental impacts of COVID-19 restrictions. In particular,

74   although observations indeed show decreases of air pollutant concentrations during COVID-19 restrictions(Fan et al., 2020;

75   Wang et al., 2021; He et al., 2020; Shi and Brasseur, 2020), the air quality improvement is much smaller than the expected

76   (Shi et al., 2021; Diamond and Wood, 2020; Yan et al., 2022). Moreover, severe haze (COVID-19 haze) still occurred in

77   northern China (Sulaymon et al., 2021; Le et al., 2020) and O_3 concentrations even showed significant increases (Zhang et

78   al., 2021; Li et al., 2020). A number of studies were conducted to explain this air quality change by analyzing the effects of

79   emission changes, meteorological variations and secondary production (Huang et al., 2021; Le et al., 2020; Hammer et al.,

80   2021; Zhao et al., 2020; Zhao et al., 2021; Sulaymon et al., 2021; Wang et al., 2020; Li et al., 2021). However, due to the

81   unknown emission changes during COVID-19 restrictions, the emission reduction scenarios that used to represent the

82   COVID-19 shutdown effects varied among different studies and did not consider the spatial and temporal heterogeneity of

83   the emission changes, leading to biases in the model simulation (Zhao et al., 2021; Li et al., 2021; Hammer et al., 2021;

84   Zheng et al., 2021) and uncertainty in the quantification of the contributions of different factors.

85   Pioneer studies by Zheng et al. (2021) and Forster et al. (2020) have derived multi-air pollutant emissions from social

86   activity data using a bottom-up method, but due to the lack of detailed social activity data, large uncertainties existed in their

87   estimates. The meteorologically and seasonally driven variability of the concentrations of air pollutants also prohibit drawing

88   fully quantitative conclusions on the changes of emissions based on observations alone (Levelt et al., 2022). The emission

89   inversion technique, which takes advantage of the chemical transport model (CTM) and real-time observations, provides an

90   attractive way to estimate the sector-specific and space-based emission changes during COVID-19 restrictions (Levelt et al.,

91   2022), as shown in Zhang et al. (2020), Zhang et al. (2021), Feng et al. (2020) and Hu et al. (2022). However, these studies

92   only inversed the emissions of single species (e.g., NO_x and SO_2) without insights into multiple species. In view of this

93   discrepancy, in this study we developed a multi-air pollutant inversion system to simultaneously estimated the multi-air

94   pollutant emissions in China, including NO_x, SO_2, CO, $PM_{2.5}$ and PM_{10}, during the COVID-19 restrictions using an ensemble

95   Kalman filter (EnKF) and surface observations from the China National Environmental Monitoring Centre (CNEMC).

96   Subsequently, the inversed emission inventory was used to quantify the contributions of emission changes versus

97   meteorology variations to the changes in $PM_{2.5}$ and O_3 concentrations over the NCP region during the COVID-19 restrictions.

98   **2 Method and data**

99   We developed a high-resolution multi-air pollutant inversion system to estimate the daily emissions of NO_x, SO_2, CO,

100 $PM_{2.5}$ and PM_{10} in China during the COVID-19 restrictions. This system uses the NAQPMS (Nested Air Quality Prediction

101 Modelling System) model as the forecast model and the EnKF coupled with the state argumentation method as the inversion
102 method. It has the capabilities of simultaneous inversion of multi-air pollutant emissions at high temporal (daily) and spatial
103 (15km) resolutions. An iteration inversion scheme was also developed in this study to address the large biases in the a priori
104 emissions. In the following sections, we briefly introduce each component of the inversion system.

2.1 Chemical transport model and its configuration

106 The NAQPMS model was used as the forecast model to represent the atmospheric chemistry in this study, which has
107 been used in previous inversion studies (Tang et al., 2011; Tang et al., 2013; Kong et al., 2019; Wu et al., 2020), where
108 detailed descriptions of NAQPMS are available. The Weather Research and Forecasting Model (WRF)(Skamarock, 2008) is
109 used to provide the meteorological inputs to the NAQPMS model.

110 Figure 1 shows the modelling domain of this study with a high horizontal resolution of 15 km. The a priori emission
111 inventory used in this study includes monthly anthropogenic emissions from the HTAP_v2.2 emission inventory for the base
112 year of 2010 (Janssens-Maenhout et al., 2015), biomass burning emissions from the Global Fire Emissions Data base (GFED)
113 version 4 (Randerson et al., 2017; Van Der Werf et al., 2010), biogenic volatile organic compound (BVOC) emissions from
114 MEGAN-MACC (Sindelarova et al., 2014), marine volatile organic compound emissions from the POET database (Granier
115 et al., 2005), soil NO_x emissions from the Regional Emission inventory in Asia (Yan et al., 2003) and lightning NO_x
116 emissions from Price et al. (1997). Chemical top and boundary conditions were provided by the global CTM MOZART
117 (Model for Ozone and Related Chemical Tracers) (Brasseur et al., 1998; Hauglustaine et al., 1998). We assumed no monthly
118 variations in the a priori emission inventory and used January's emission inventory for the whole simulation period so that
119 the emission variation was solely derived from the surface observations. A two-week free run of NAQPMS was conducted
120 as a spin-up time. For each day's meteorological simulation, a 36-h free run of WRF was conducted, of which the first 12-h
121 simulation was a spin-up run and the next 24-h simulation provided the meteorological inputs to NAQPMS. Initial and
122 boundary conditions for the meteorological simulation were provided by the National Center for Atmospheric
123 Research/National Center for Environment Prediction (NCAR/NCEP) 1°×1° reanalysis data. Evaluation results for the
124 WRF simulation are available in Text S1 in Supplement.

2.2 Surface Observations

126 The hourly concentrations of NO_2, SO_2, CO, $PM_{2.5}$ and PM_{10} from CNEMC were used in this study to estimate the
127 emissions during COVID-19. The spatial distributions of these observation sites are shown in Fig. 1, which contains 1436
128 observation sites covering most regions of China. Before assimilation, outliers of observations were first filtered out using
129 the automatic outlier detection method developed by Wu et al. (2018) to prevent the adverse effects of the outliers on data
130 assimilation. Then, the hourly concentrations were averaged to the daily values for the inversions of daily emissions.

131 The observation error is one of the key inputs to the data assimilation, which together with the background error
132 determine the relative weights of the observation and background values on the analysis. The observation error includes

133   measurement error and representativeness error. The measurement error of each species was designated according to the

134   officially released documents of the Chinese Ministry of Ecology and Environmental Protection (HJ 193-2013 and HJ 654-

135   2013, available at http://www.cnemc.cn/jcgf/dqhj/, last accessed on 22 Oct 2022), which is 5% for $PM_{2.5}$ and PM_{10} and 2%

136   for SO_2, NO_2 and CO. A representativeness error arises from the different spatial scales that the discrete observation data and

137   model simulation represent, which was estimated based on the previous study by Li et al. (2019) and Kong et al. (2021).

2.3 Inversion estimation scheme

139   The EnKF coupled with the state augmentation method was used in this study to constrain the emissions of multiple

140   species. EnKF is an advanced data assimilation method proposed by Evensen (1994) that features representation of the

141   uncertainties of the model state by a stochastic ensemble of model realizations. It is easily implemented and can update the

142   state variable by using the flow-dependent background error covariance matrix. In the state augmentation method, the

143   emissions of different species, together with the concentrations of related species, were treated as the state variable and were

144   updated according to the relationship between the emissions and concentrations of related species.

145   Since the source emission data over mainland China in HTAP_v2.2 inventory is obtained from the MIX inventory (Li et

146   al., 2017b), uncertainties of emissions of different species, including PMF, PMC, BC, OC, NO_x, CO, SO_2, NH_3 and NMVOC

147   (nonmethane volatile organic compounds), were obtained from Li et al. (2017b) and Streets et al. (2003), which were

148   represented by an ensemble of perturbed emissions generated by multiplying the a priori emissions with a perturbation factor

149    $\beta_{i,s}$:

150    $$\boldsymbol{E}_{i,s} = \boldsymbol{\beta}_{i,s} \circ \boldsymbol{E}_s^p, \ i = 1,2,\cdots N_{ens} \tag{1}$$

151   where $E_{i,s}$ represents the vector of the ith member of perturbed emissions for species s, E_s^p represents the a priori emissions

152   for this species, $\circ$ denotes the schur product and N_{ens} denotes the ensemble size. Thus, the adjustment of emissions is

153   equivalent to the adjustment of perturbation factors.

154   Considering that emission uncertainty is the major contributor to the uncertainties in air quality modelling, especially

155   during the COVID-19 period when emissions changed rapidly, uncertainties in chemical variables were obtained through

156   ensemble simulations driven by perturbed emissions. The ensemble size was chosen as 50 to maintain the balance between

157   the filter performance and computational cost. After the ensemble simulations, emissions of multiple species were updated

158   using a deterministic form of EnKF (DEnKF) proposed by Sakov and Oke (2008), which is formulated by

159    $$\overline{x^a} = \overline{x^b} + \mathbf{P}_e^b \mathbf{H}^T \left(\mathbf{H}\mathbf{P}_e^b \mathbf{H}^T + \mathbf{R}\right)^{-1} \left(\boldsymbol{y}^o - \mathbf{H}\overline{x^b}\right) \tag{2}$$

160    $$\overline{x^b} = \frac{1}{N}\sum_{i=1}^N x_i^b \ ; X_i^b = x_i^b - \overline{x^b} \tag{3}$$

161    $$\mathbf{P}_e^b = \frac{1}{N-1}\sum_{i=1}^N X_i^b \left(X_i^b\right)^T \tag{4}$$

162   where x denotes the state variables; b the background state (a priori); a the analysis state (posteriori); $\mathbf{P}_e^b$ the ensemble-

163   estimated background error covariance matrix and N the ensemble size. $\boldsymbol{y}^o$ represents the vector of observations with an

164 error covariance matrix of **R**. **H** is the linear observational operator that maps the m-dimensional state vector $\boldsymbol{x}$ to a p-

165 (number of observations) dimensional observational vector ($\mathbf{H}\overline{\boldsymbol{x^b}}$). The state variables were defined as follows according to

166 state augmentation method during the assimilation:

167 $\boldsymbol{x}_i = [\boldsymbol{c}_i, \ \boldsymbol{\beta}_i]^T, i = 1,2,\cdots N_{ens}$ (5)

168 $\boldsymbol{c}_i = [\boldsymbol{PM_{2.5}}, \boldsymbol{PM_{10-2.5}}, \ \boldsymbol{NO_2}, \boldsymbol{SO_2}, \boldsymbol{CO}]_i$ (6)

169 $\boldsymbol{\beta}_i = [\boldsymbol{\beta_{PMF}}, \boldsymbol{\beta_{BC}}, \ \boldsymbol{\beta_{OC}}, \ \boldsymbol{\beta_{PMC}}, \boldsymbol{\beta_{NO_x}}, \boldsymbol{\beta_{SO_2}}, \boldsymbol{\beta_{CO}}]_i$ (7)

170 where $\boldsymbol{x}_i$ represents the ith member of the assimilated state variable, which consists of the fields of chemical variables $\boldsymbol{c}_i$

171 and emission perturbation factors $\boldsymbol{\beta}_i$. Detailed descriptions of the model state variables are summarized in Table 1. The use

172 of PM$_{10-2.5}$ (PM$_{10}$ minus PM$_{2.5}$) values aims to avoid the potential cross-correlations between PM$_{2.5}$ and PM$_{10}$ (Peng et al.,

173 2018; Ma et al., 2019). Moreover, to prevent spurious correlations between non- or weakly related variables, similar to Ma et

174 al. (2019) and Miyazaki et al. (2012), state variable localization was used during assimilation, with observations of one

175 particular species only allowing us to update the emissions of the same species. The corresponding relationship between the

176 chemical observations and adjusted emissions is summarized in Table 1. The PM$_{2.5}$ observations were one exception and

177 were used to update the emissions of PMF (fine mode unspeciated aerosol), BC (black carbon) and OC (organic carbon)

178 since the observations of speciated PM$_{2.5}$ were not available in this study. The lack of speciated PM$_{2.5}$ observations may lead

179 to uncertainties in the estimated emissions of PMF, BC and OC. Therefore, we only analyzed the emissions of PM$_{2.5,}$ which

180 were the sum of the emissions of these three species. Similarly, only PM$_{10}$ emissions were analyzed in this study, which

181 includes the emissions of PM$_{2.5}$ and PMC (coarse mode unspeciated aerosol).

182   Due to the strict control measures implemented during the last decades, the emissions in China decreased dramatically

183 from 2010 to 2020, especially for SO$_2$. Thus, there are large biases in the a priori estimates of emissions in China (Zheng et

184 al., 2018), which would lead to incomplete adjustments of a priori emissions and degrade the performance of assimilation.

185 Therefore, an iteration inversion scheme was developed in this study to address the large biases of SO$_2$ emissions. As

186 illustrated in Fig. 2, the main idea of the iteration inversion scheme is to update the ensemble mean of the state variable using

187 the inversion results of the kth iteration and corresponding simulations. The state variable used in the $(k + 1)th$ inversions

188 is written as follows:

189 $\boldsymbol{x}_i^{k+1} = \left[\boldsymbol{c}^k + \boldsymbol{c}_i^e - \overline{\boldsymbol{c}^e}, \boldsymbol{\beta}^k + \boldsymbol{\beta}_i^e - \overline{\boldsymbol{\beta}^e}\right]^T$ (8)

190 where $\boldsymbol{c}^k$ represents the simulation results using the inversed emissions of the kth iteration, $\boldsymbol{c}_i^e$ represents the ith member of

191 ensemble simulations with an ensemble mean of $\overline{\boldsymbol{c}^e}$, $\boldsymbol{\beta}^k$ represents the perturbation factors of the kth iteration, and

192 $\boldsymbol{\beta}_i^e$ represents the ith member of the ensemble of perturbation factors with a mean value of $\overline{\boldsymbol{\beta}^e}$.

193   Using this method, the biases of a priori emissions were well addressed as exemplified in Fig. 3 for SO$_2$ emissions. Due

194 to the large positive biases in the a priori SO$_2$ emissions, the model still has large positive biases (NMB = 30.9–220.5%) and

195 errors (RMSE = 8.7–23.0 µg/m^3) in simulating SO$_2$ concentration over all regions of China even after the assimilation (first

196 iteration). The biases and errors continued to decrease with the increasing of iteration times till the fourth iteration in which

197 there were no significant improvement in SO$_2$ simulations compared to those in third iteration. These results suggested that

198 the iteration inversion method used in this study can well constrain the a priori emission with large biases and, in this

199 application, conducting three iteration is enough for constraining the emission. Besides SO$_2$ emissions, the iteration inversion

200 scheme was also applied to the emissions of other species.

201   To reduce the influences of random model errors (e.g., errors in meteorological inputs) on the estimation of the

202 variation in emissions, a 15-day running average was performed on our daily inversion results after the inversion estimation.

203 It should be noted that the COVID-19 restrictions were initiated during the Spring Festival of China, which would also

204 influence the air pollutant emissions in China. However, the inversion method used in this study did not differentiate the

205 contributions of the Spring Festival from the COVID-19 restrictions. Similarly, the effects of natural emission changes were

206 not differentiated in this study, which would lead to uncertainty in quantifying the effects of the COVID-19 restrictions on

207 air pollutant emissions.

2.4 Quantification of the effects of emission changes and meteorological variations

209   In previous studies, the meteorological-induced (MI) changes were usually determined by the CTM with a fixed

210 emission input setting and a varying meteorological input. Then, the difference between the MI changes and total changes in

211 air pollutant concentrations is defined as emission-induced (EI) changes. Another approach to estimate EI changes is to

212 perform simulations with a fixed meteorological input setting and varying emission inputs. Then, the MI changes are defined

213 as the difference between EI changes and total changes in air pollutant concentrations. Due to the nonlinear effects of

214 atmospheric chemical systems, these two methods yield different results. Thus, both methods were used in this study to

215 account for the nonlinear effects. The averaged results of these two methods are used to represent the impacts of emission

216 changes and meteorological variation on the air quality changes during the COVID-19 restrictions. In total, three scenario

217 experiments were designed based on our inversion results. The first scenario simulation used the varying meteorological and

218 emission inputs from the P1 to P2 period, which represents the real-world scenario and is used to estimate the total changes

219 in air pollutant concentrations induced by emissions and meteorological changes from the P1 to P2 period (BASE scenario).

220 The second scenario experiment used the varying meteorological inputs but replaced the emissions during the P2 period with

221 those during the P1 period, which was used to estimate the MI changes using the first method (MET change scenario). The

222 third scenario experiment used the varying emissions input and replaced the meteorological input during the P2 period with

223 that during the P1 period, which was used to estimate the EI changes using the second method (EMIS change scenario).

224 Based on the first method, the MI and EI changes can be estimated as follows:

225 $MI_{MET\ change\ scenario} = conc_{p2,MET\ change\ scenario} - conc_{p1,MET\ change\ scenario}$ (9)

226 $EI_{MET\ change\ scenario} = conc_{p2,BASE\ scenario} - conc_{p1,BASE\ scenario} - MI_{MET\ change\ scenario}$ (10)

227 where $MI_{MET\ change\ scenario}$ represents the MI changes estimated based on the results from the MET change scenario,

228 $conc_{p1,MET\ change\ scenario}$ and $conc_{p2,MET\ change\ scenario}$ represent the averaged concentrations of air pollutants during the

229   P1 and P2 periods under the MET change scenario, $EI_{MET\ change\ scenario}$ represents the EI changes estimated based on the

230   results from the MET change scenario, and $conc_{p1,BASE\ scenario}$, $conc_{p2,BASE\ scenario}$ respectively represent the averaged

231   concentrations of air pollutants during the P1 and P2 periods under the BASE scenario. Similarly, the MI and EI changes

232   estimated based on the second method are formulated as follows:

233   $EI_{EMIS\ change\ scenario} = conc_{p2,EMIS\ change\ scenario} - conc_{p1,EMIS\ change\ scenario}$ (11)

234   $MI_{EMIS\ change\ scenario} = conc_{p2,BASE\ scenario} - conc_{p1,BASE\ scenario} - EI_{EMIS\ change\ scenario}$ (12)

235   Then, the estimations from these two methods are averaged to estimate the contributions of meteorological change and

236   emission change to the changes in PM$_{2.5}$ and O$_3$ concentrations during the COVID-19 lockdown:

237   $MI = (MI_{EMIS\ change\ scenario} + MI_{MET\ change\ scenario})/2$ (13)

238   $EI = (EI_{EMIS\ change\ scenario} + EI_{MET\ change\ scenario})/2$ (14)

239   $contri_{met} = \frac{MI}{MI+EI} \times 100$ (15)

240   $contri_{emis} = \frac{EI}{MI+EI} \times 100$ (16)

241   where $contri_{met}$ and $contri_{emis}$ represent the relative contributions (%) of the meteorological variations and emission

242   changes to the changes in air pollutant concentrations.

243   **3 Results**

244   We estimated the multi-air pollutant emissions from 1 Jan to 29 Feb 2020 when the pandemic was at its most serious,

245   and the effects of the COVID-19 restrictions were most profound in China. According to different control phases of COVID-

246   19 and the timing of the Chinese Lunar New Year, the whole time period was divided into three periods: before lockdown

247   (P1, January 1-20), lockdown (P2, January 21-February 9) and after back-to-work day (P3, February 10-29) to better

248   characterize the emission changes during the COVID-19 restrictions. Additionally, we analyzed the emission changes in

249   different regions of China, including the North China Plain (NCP), Northeast China (NE), Southeast China (SE), Southwest

250   China (SW), Northwest China (NW) and Central regions (defined in Fig. 1) to investigate the responses of emissions to the

251   COVID-19 restrictions in different regions.

252   **3.1 Validation of the inversion results**

253   We firstly validate our inversion system by using a cross-validation, in which 20% of observation sites were withheld

254   from the emission inversion and used as the validation datasets. Figure S1–6 showed the concentrations of different air

255   pollutants in China from 1st Jan to 29th Feb 2020 obtained from observations at validation sites and simulations using a priori

256   and a posteriori emission. Commonly used statistical evaluation indices, including correlation coefficient (R), mean bias

257   error (MBE), normalized mean bias (NMB) and root of mean square error (RMSE) are summarized in Table S1. The

258   validation results suggest that the posteriori simulation agreed well with the observed concentrations for all species. The

259   large biases in the a priori simulation of PM$_{2.5}$, PM$_{10}$, SO$_2$ and CO were almost completely removed in the a posteriori

260   simulation with NMB about -3.9–15.7% for PM$_{2.5}$, -3.1–11.6% for PM$_{10}$, -12.6–5.3% for NO$_2$, -9.5–6.2% for SO$_2$ and -10–

261   7.6% for CO (Table S4). RMSE values were also significantly reduced in the a posteriori simulation which were 9.1–

262   32.2μg/m^3 for PM$_{2.5}$, 12.6–42.4μg/m^3 for PM$_{10}$, 5.1–12.3μg/m^3 for NO$_2$, 1.2–5.6μg/m^3 for SO$_2$ and 0.10–0.46mg/m^3 for

263   CO. Moreover, the inversion emission considerably improved the fit to the observed time evolution of air pollutants'

264   concentrations. The R values were improved for all species in the a posteriori simulation that were up to 0.74–0.94 for PM$_{2.5}$,

265   0.63 – 0.92 for PM$_{10}$, 0.76–0.94 for NO$_2$, 0.23–0.79 for SO$_2$ and 0.63–0.92 for CO. These results suggest that our inversion

266   results have excellent performance in representing the magnitude and variation of these species' emission in China during

267   COVID-19 restrictions. Model performance in simulating O$_3$ concentration is relatively poor compared to other species

268   although improvement was remarkable in NCP, NE and SE regions. This would be due to that the emission of volatile

269   organic compounds, another important precursor for O$_3$, were not constrained in this study.

3.2 Emission changes of multi-species during COVID-19 restrictions

3.2.1 Unbalanced emission changes between NO$_x$ and other species

272   The control of COVID-19 began on 23rd January when the Chinese government declared the first level of national

273   responses to public health emergencies, one day before the 2020 Chinese New Year Eve. Figure 4 shows the time evolution

274   of the normalized emission anomaly for different species in China from 1st January to 29th February. The temporal variation

275   in the emission varied largely between NOx and other species. Due to the combined effects of the Spring Festival and

276   COVID-19 lockdown, NO$_x$ emissions decreased continuously at the beginning of January until approximately one week after

277   the implementation of the COVID-19 lockdown, with estimated decreases in NO$_x$ emissions of up to 46.7% from the P1 to

278   P2 period (Table 2). Subsequently, the NO$_x$ emissions stabilized with small fluctuations until the official back-to-work day

279   when the NOx emissions began to increase due to easing of the control measures and the resumption of business. According

280   to inversion estimation, NO$_x$ emissions recovered by 6.5% during the P3 period. These results indicate that the temporal

281   variation in our estimated NO$_x$ emissions agreed well with the timing of the Spring Festival and different control stages of

282   COVID-19. However, for other species (i.e., PM$_{2.5}$, PM$_{10}$, SO$_2$ and CO), although their emissions generally decreased from

283   1st January to the end of the 2020 Spring Festival holiday, they showed much smaller reductions than the NO$_x$ emissions.

284   The emission reduction for these species was only approximately 7.9-12.1% (Table 2). This is consistent with the inversion

285   results by Hu et al. (2022) who found that SO$_2$ emissions in China decreased only by 9.2% during COVID-19 lockdown. In

286   addition, the emissions of these species quickly rebounded to their normal level just one week after the end of the Spring

287   Festival holiday. As estimated by our inversion results, the SO$_2$ emissions recovered by 7.2% during the P3 period, which

288   was only 2.5% lower than that during the P1 period. The PM$_{2.5}$ and PM$_{10}$ emissions during the P3 period were 3.3% and 43.6%

289   higher, respectively, than those during the P1 period.

290   Similar results were found in different regions of China (Fig. 5 and Table 3), where the NO$_x$ emissions decreased much

291   more than other species. In addition, unlike the uniform decreases in NO$_x$ emissions in different regions of China (~45%),

292   there was apparent spatial heterogeneity in the emission changes in PM$_{2.5}$, PM$_{10}$, SO$_2$ and CO (Table 3 and Fig. 6). For

293   example, from the P1 to P2 period, the PM$_{2.5}$ emissions decreased by over 20% in the Central region but only by 8.8% in the

294   NE region. The PM$_{2.5}$ emissions even increased by 5.5% in the NCP region, possible due to the increased emissions from

295   heating and fireworks (Dai et al., 2020). The SW and central regions exhibited relatively larger emission reductions for these

296   species (Fig. 5 and Table 3) by 12.6–25.9% and 10.6–23.7%, respectively. The emission rebound during the P3 period was

297   more prominent in the SE, central and SW regions (Fig. 5 and Fig. 7), where emissions recovered by 6.0–16.4% for NO$_x$,

298   7.5–19.8% for SO$_2$, 7.4–13.1% for CO, 12.3–47.7% for PM$_{2.5}$ and 28.6–135.9% for PM$_{10}$ (Table 3). This result is consistent

299   with the earlier degradation of the response level to the COVID-19 virus (from the first level to the second or third level)

300   over these regions (Table S2). In contrast, there were decreases in emissions in the NCP, NE and NW regions. PM$_{2.5}$

301   emissions were reduced by 9.9% in the NCP region and by 19.2% in the NE region from the P2 to P3 period (Table 3).

302   Moreover, we found that the PM$_{10}$ emissions surged in the NW and central regions, where the PM$_{10}$ emissions during the P3

303   period were almost two times larger than those during the P2 period (Table 3). However, this finding may be related to the

304   enhanced sandstorms over these two regions rather than the effects of returning to work, which demonstrates the necessity to

305   consider changes in natural emissions during COVID-19 restrictions. Thus, to reduce the effects of natural emissions on our

306   findings, the same analysis was performed for the emissions over east China (Fig. S7) where emissions were dominated by

307   anthropogenic sources, which shows consistent results with the findings above (Fig. S8 and Table S3).

3.2.3 Explanations for the emission changes during COVID-19 restrictions

309   Two explanations may help clarify the unbalanced emission changes between NO$_x$ and other species. First, the COVID-

310   19 lockdown policy has led to dramatic decreases in transportation activities throughout China; however, as shown in Fig. 4,

311   the relative contributions of the transportation sector to the emissions of SO$_2$ (2.4%), CO (18.5%), PM$_{2.5}$ (6.1%) and PM$_{10}$

312   (4.7%) are much smaller than those for NO$_x$ emissions (34.3%) (Zheng et al., 2018; Li et al., 2017a). Thus, the reduction in

313   traffic activities can only substantially decrease NO$_x$ emissions. Reductions in CO emissions were relatively larger than those

314   for SO$_2$ and PM$_{2.5}$ emissions, which is consistent with the relatively larger contributions of the transportation sector to CO

315   emissions. PM$_{10}$ emissions showed the largest reductions among these four species, which is related in part to the reduced

316   dust emissions due to shutting down of construction sites during the lockdown period (Li et al., 2020). Second, as shown in

317   Fig. 4, the industrial and residential sectors are the major contributors to the anthropogenic emissions of SO$_2$, CO, PM$_{2.5}$ and

318   PM$_{10}$ in China, together contributing 77.6%, 78.3%, 86.5% and 86.3%, respectively, to their total emissions. The much

319   smaller reductions of these species' emissions were thus in line with the fact that there were no intentional restrictions on

320   heavy industry during the COVID-19 restrictions. A large number of non-interruptible processes, such as steel, glass, coke,

321   refractory, petrochemical, electric power, and especially heating, cannot be stopped during the COVID-19 lockdown.

322   According to statistical data from the National Bureau of Statistics of China (Fig. S9), the industrial and power sectors did

355   The emission changes contributed slightly to the $PM_{2.5}$ reductions over the NCP region (8.6%). This is because, on the

356   one hand, the large reductions in NO_x emissions (by 44.4%) only reduced nitrate by approximately 10–30% due to the

357   nonlinear effects of chemical reactions (Fig. 11), and on the other hand, the emissions of primary $PM_{2.5}$ and its precursors

358   from other sectors changed little during the COVID-19 restrictions (Table 3). The emission changes contributed more to the

359   increased $PM_{2.5}$ concentrations over the Beijing area (42.1%). This is mainly associated with the increases in primary $PM_{2.5}$

360   emissions around the Beijing area, as seen in Fig. 6, possibly due to the increased emissions from firework during the Spring

361   Festival over the rural area of Beijing (Dai et al., 2020). Therefore, our results suggested that the unexpected COVID-19

362   haze was mainly driven by unfavorable meteorological conditions together with small changes or even increases in primary

363     $PM_{2.5}$ emissions. This finding is in line with previous results of Le et al. (2020) but different from those of Huang et al.

364   (2021), who suggested that enhanced secondary aerosol formation was the main driver of severe haze during the COVID-19

365   restrictions. To investigate it, we further analyzed the changes in the concentrations of secondary inorganic aerosols (SIAs).

366   First, we evaluated our model results against the observed SIA concentrations, which showed that the model results using

367   our inversion emissions well reproduced the observed concentrations of SIAs over the NCP region (Fig. 12) with mean bias

368   (MB) ranging from -5.14 to 5.45 μg/m^3 and correlation coefficient (R) ranging from 0.59 to 0.80. The observed increases in

369   SIA concentrations over the Beijing area, especially for sulfate concentrations, were also captured in our simulations (Fig.

370   11), although underestimation occurred due to the uncertainty in simulating SIA concentrations. Through sensitivity

371   experiments, we found that the increases in SIA concentrations were still driven by meteorological variations (Fig. 13). In

372   fact, the emission reductions only led to a 10% decrease in SIA concentrations over the NCP region. This finding suggests

373   that the enhanced secondary aerosol formation was likely mainly driven by the unfavorable meteorological conditions

374   associated with higher temperature and relative humidity instead of the emission reductions during the lockdown period.

375   This is in line with the observation evidences from Ma, T et al (2022) who emphasized that the increased temperature and

376   relative humidity promoted the formation of secondary pollutants during the COVID-19 restrictions.

377   In terms of O_3 concentrations, the emission changes subsequently became the dominant contributor to the O_3 increases

378   by more than 100% in the Beijing area and by 96.0% over the NCP region. This result is mainly because the lockdown

379   period occurred in midwinter when photochemical O_3 formation was minimal; thus, the large increase in O_3 is expected

380   solely from the effect of the reduced titration reaction associated with the large reductions in NO_x emissions. Although the

381   higher temperature and slower wind speed during the lockdown period were favorable for the increases in O_3 concentrations,

382   their contributions were much smaller than those of emission changes (Fig. 9). These results suggested that control measures,

383   such as COVID-19 restrictions, were inefficient for air pollution mitigation in China considering the high economic cost of

384   the COVID-19 restrictions.

385 **4 Conclusions**

386 The COVID-19 pandemic is an unprecedented event that significantly influenced the social activity and associated

387 emissions of air pollutants. Our results provide a quantitative assessment of the influences of COVID-19 restrictions on

388 multi-air pollutant emissions in China. Otherwise, understanding of the relationship between air quality and human activities

389 may be biased. The inversion results provide important evidences that the COVID-19 lockdown policy was largely a traffic

390 control measure with substantially reducing impacts on NO_x emissions but much smaller influences on the emissions of other

391 species and other sectors. Traffic control has widely been considered to be the normal protocol in implementing regulations

392 in many cities of China, but its effectiveness on air pollution control is still disputed (Han and Naeher, 2006; Zhang et al.,

393 2007; Chen et al., 2021; Cai and Xie, 2011; Chowdhury et al., 2017; Li et al., 2017c). Thus, the COVID-19 restrictions

394 provided us with a real nationwide traffic control scenario to investigate the effectiveness of traffic control on the mitigation

395 of air pollution in China. The results suggested that traffic control as a separate pollution control measure has limited effects

396 on the coordinated control of high concentrations of O_3 and $PM_{2.5}$ under the current air pollution conditions in China. In this

397 case, the $PM_{2.5}$ concentrations were slightly reduced, while leading to substantial increases in O_3 concentrations. Severe haze

398 was also not avoided during the COVID-19 restrictions due to unbalanced emission changes from other sectors and

399 unfavorable meteorological conditions. China is now facing major challenges in both controlling $PM_{2.5}$ and controlling

400 emerging O_3 pollution. The tragic COVID-19 pandemic has revealed the limitation of the road traffic control measure in the

401 coordinated control of $PM_{2.5}$ and O_3. More comprehensive regulations for multiple precursors from different sectors are

402 required in the future to address O_3 and $PM_{2.5}$ pollution in China.

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416 **Tables**

417 **Table 1. Corresponding relationship between the chemical observations and adjusted emissions**

Species	Descriptions	Observations that used for inversions of this species
BC	black carbon	$PM_{2.5}$
OC	organic carbon	$PM_{2.5}$
PMF	fine mode unspeciated aerosol	$PM_{2.5}$
PMC	coarse mode unspeciated aerosol	$PM_{10} - PM_{2.5}$
NO_x	nitrogen oxide	NO_2
SO_2	sulfur dioxide	SO_2
CO	carbon monoxide	CO

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420 **Table 2. Inversion estimated emissions of different air pollutants in China and their changes between different periods during**
421 **COVID-19.**

	NO_x	SO_2	CO	$PM_{2.5}$	PM_{10}
P1 (Gg/day)	72.9	23.8	1160.2	44.5	75.5
P2 (Gg/day)	41.9	21.5	1037.4	40.9	66.4
P3 (Gg/day)	44.8	23.2	1078.2	45.9	108.4
(P2-P1)/P1	-42.5%	-9.7%	-10.6%	-7.9%	-12.1%
(P3-P2)/P1	3.9%	7.2%	3.6%	11.2%	55.7%
(P3-P1)/P1	-38.6%	-2.5%	-7.0%	3.3%	43.6%

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Table 3. Inversion estimated emission changes of different air pollutants over different regions in China between different periods during COVID-19 restrictions

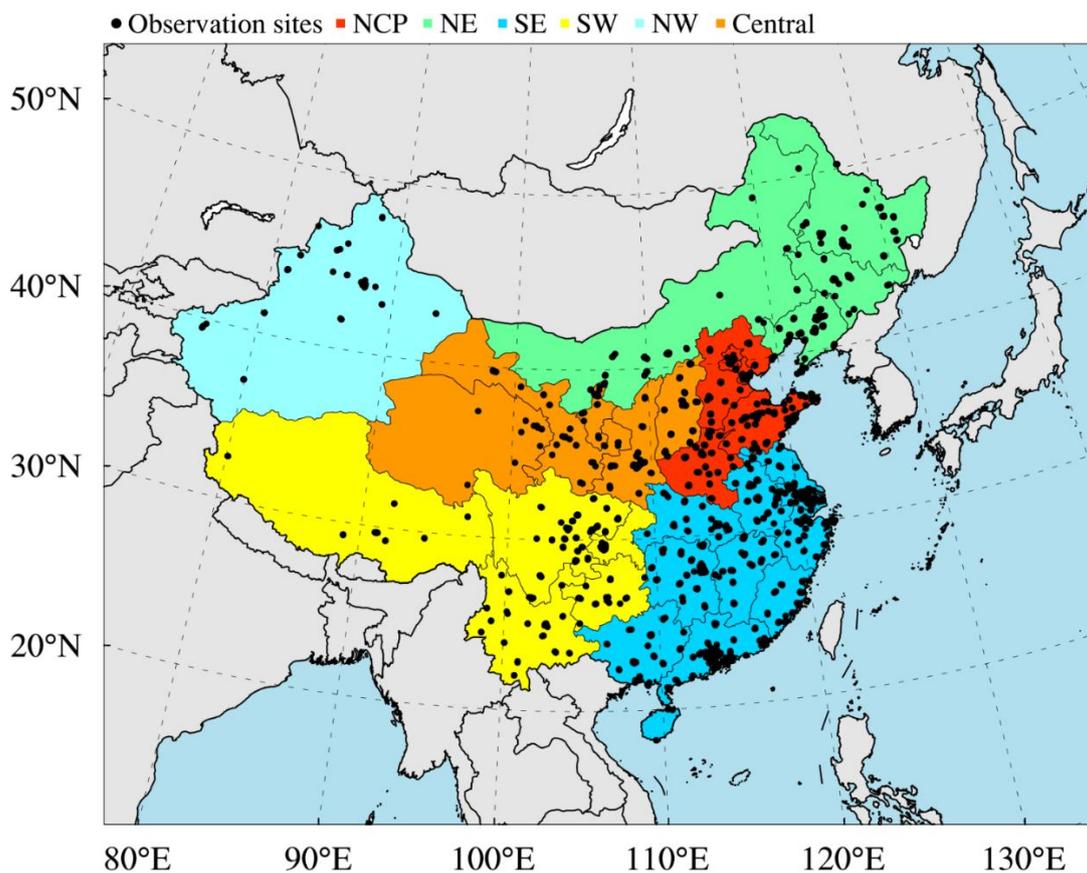
	NO$_x$	PM$_{2.5}$	PM$_{10}$	SO$_2$	CO
NCP					
(P2-P1)/P1	-44.4%	5.5%	2.8%	-1.6%	-4.3%
(P3-P2)/P1	-0.8%	-9.9%	31.8%	-5.9%	-10.0%
(P3-P1)/P1	-45.2%	-4.3%	34.7%	-7.5%	-14.3%
NE					
(P2-P1)/P1	-41.8%	-8.8%	-3.5%	-3.2%	-10.9%
(P3-P2)/P1	-6.0%	-19.2%	23.7%	-2.9%	-6.6%
(P3-P1)/P1	-47.8%	-28.0%	20.2%	-6.1%	-17.5%
SE					
(P2-P1)/P1	-41.4%	-9.5%	-24.4%	-19.4%	-3.5%
(P3-P2)/P1	10.2%	12.3%	28.6%	19.8%	13.1%
(P3-P1)/P1	-31.2%	2.8%	4.2%	0.3%	9.7%
SW					
(P2-P1)/P1	-43.5%	-12.6%	-25.9%	-17.5%	-23.8%
(P3-P2)/P1	6.0%	47.7%	33.1%	7.5%	7.4%
(P3-P1)/P1	-37.5%	35.1%	7.2%	-10.0%	-16.4%
NW					
(P2-P1)/P1	-38.5%	-4.0%	-8.3%	14.2%	-2.6%
(P3-P2)/P1	-21.1%	4.9%	145.3%	-4.1%	-7.2%
(P3-P1)/P1	-59.6%	0.9%	136.9%	10.1%	-9.8%
Central					
(P2-P1)/P1	-43.8%	-23.7%	-15.7%	-10.6%	-17.4%
(P3-P2)/P1	16.4%	24.4%	135.9%	18.5%	8.4%
(P3-P1)/P1	-27.4%	0.7%	120.3%	7.9%	-9.0%

Figures

Figure 1: Modeling domain of the ensemble simulation overlay the distributions of observation sites from CNEMC. Different colours denote the different regions in mainland of China, namely North China Plain (NCP), Northeast China (NE), Southwest China (SW), Southeast China (SE), Northwest China (NW) and Central.

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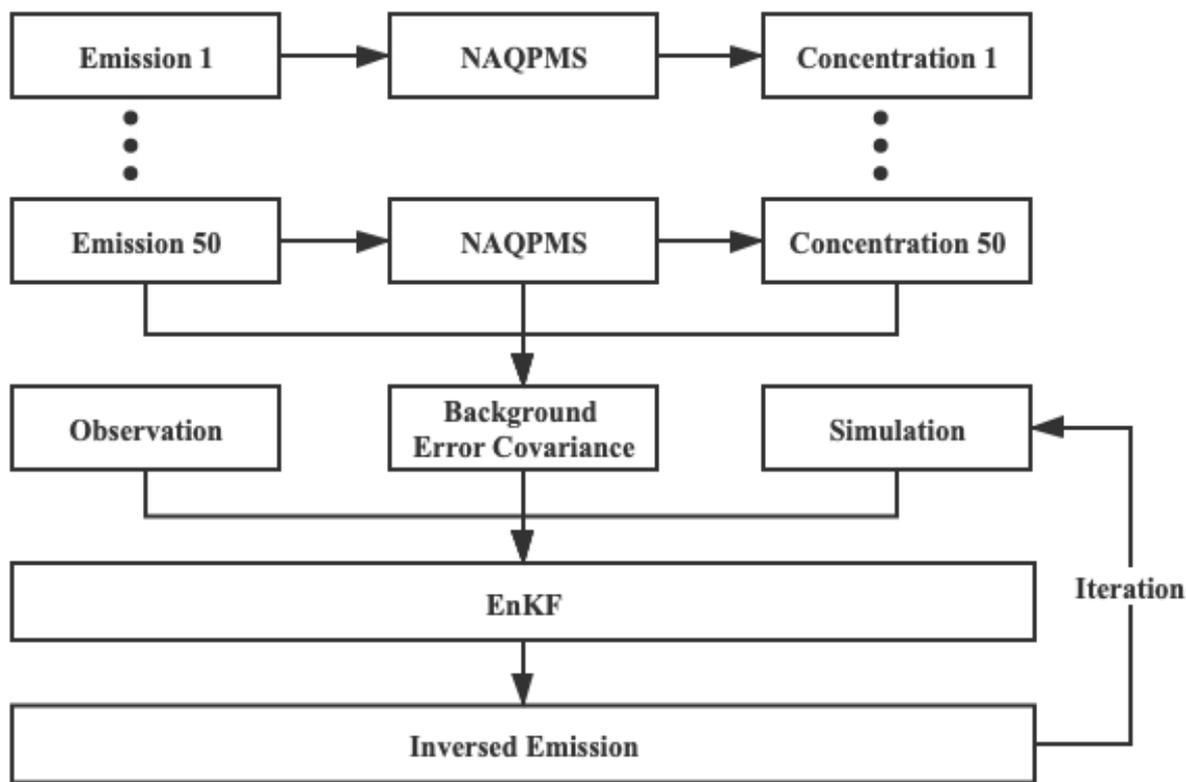
Figure 2: Illustration of the iteration inversion scheme used in this study.

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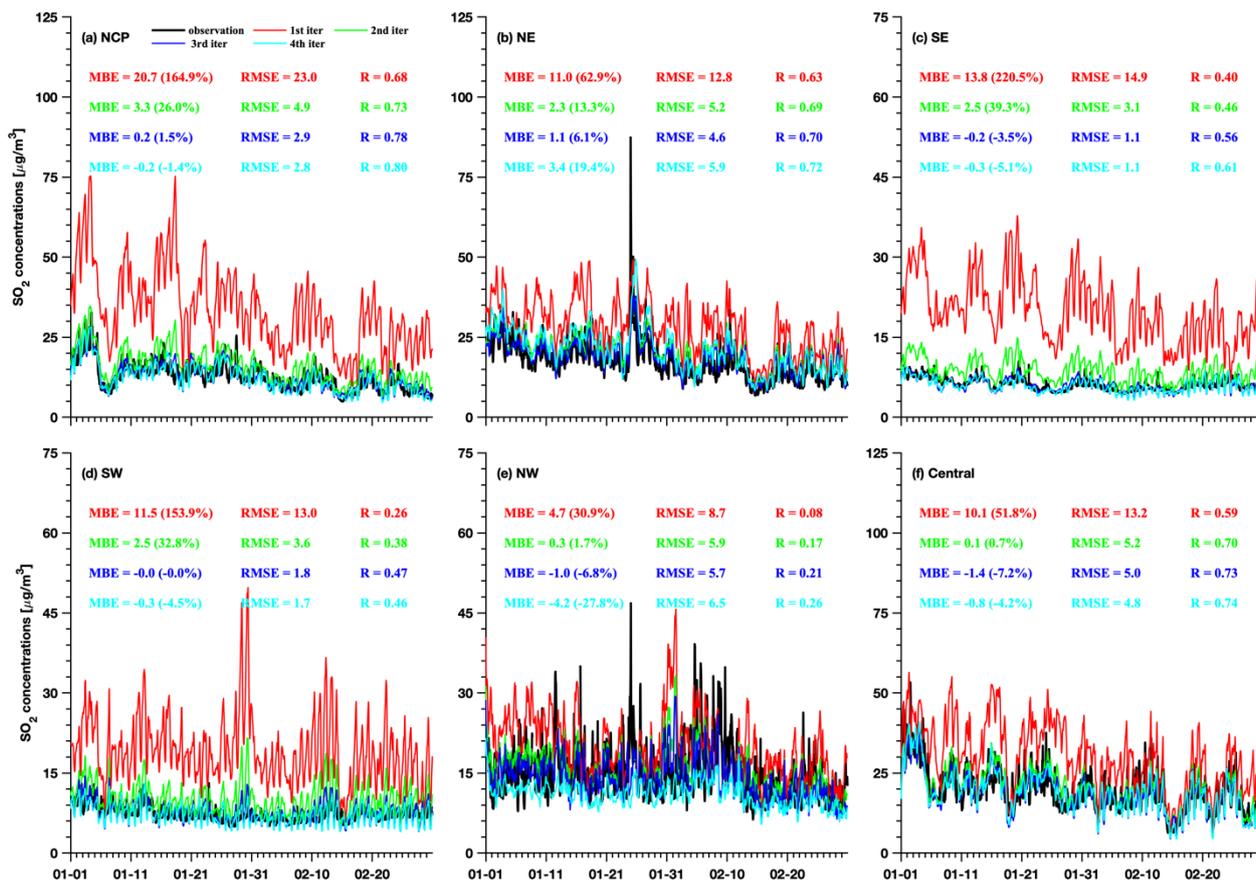
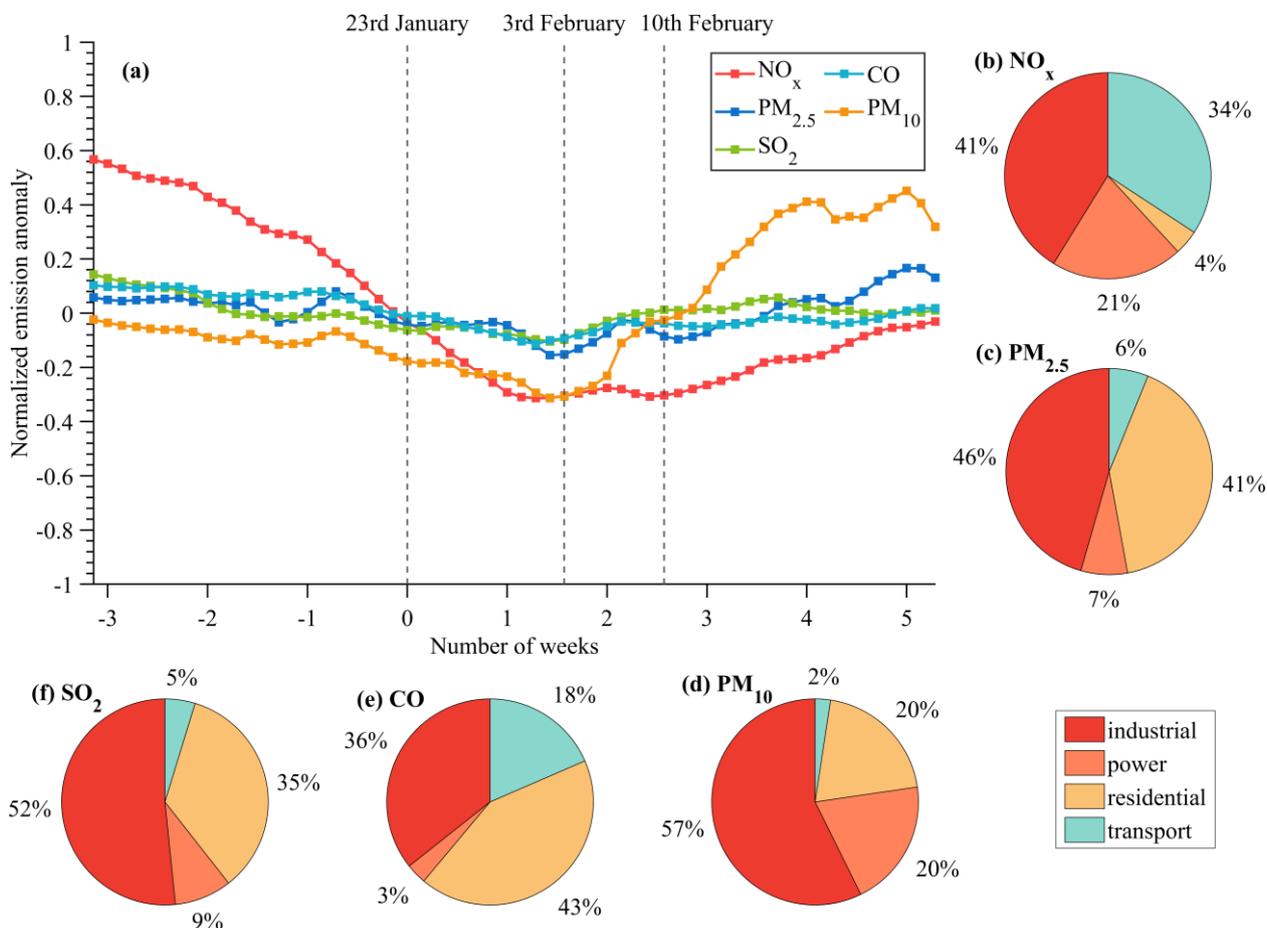


Figure 3: Comparisons of the observed and simulated mean SO2 concentrations using emissions of different iteration time at validation sites over (a) NCP region, (b) NE region, (c) SE region, (d) SW region, (e) NW region and (f) Central region.

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445 **Figure 4: (a) Time series of normalized emission anomalies estimated by inversion results for different species in China from 1st**
446 **January to 29th February 2020, and (b-f) Relative contributions of different sectors to the total anthropogenic emissions of NO$_x$,**
447 **PM2.5, PM10, CO and SO2 obtained from Zheng et al. (2018). The normalized emission anomaly is calculated by the emission**
448 **anomaly divided by the average emissions during the whole period.**
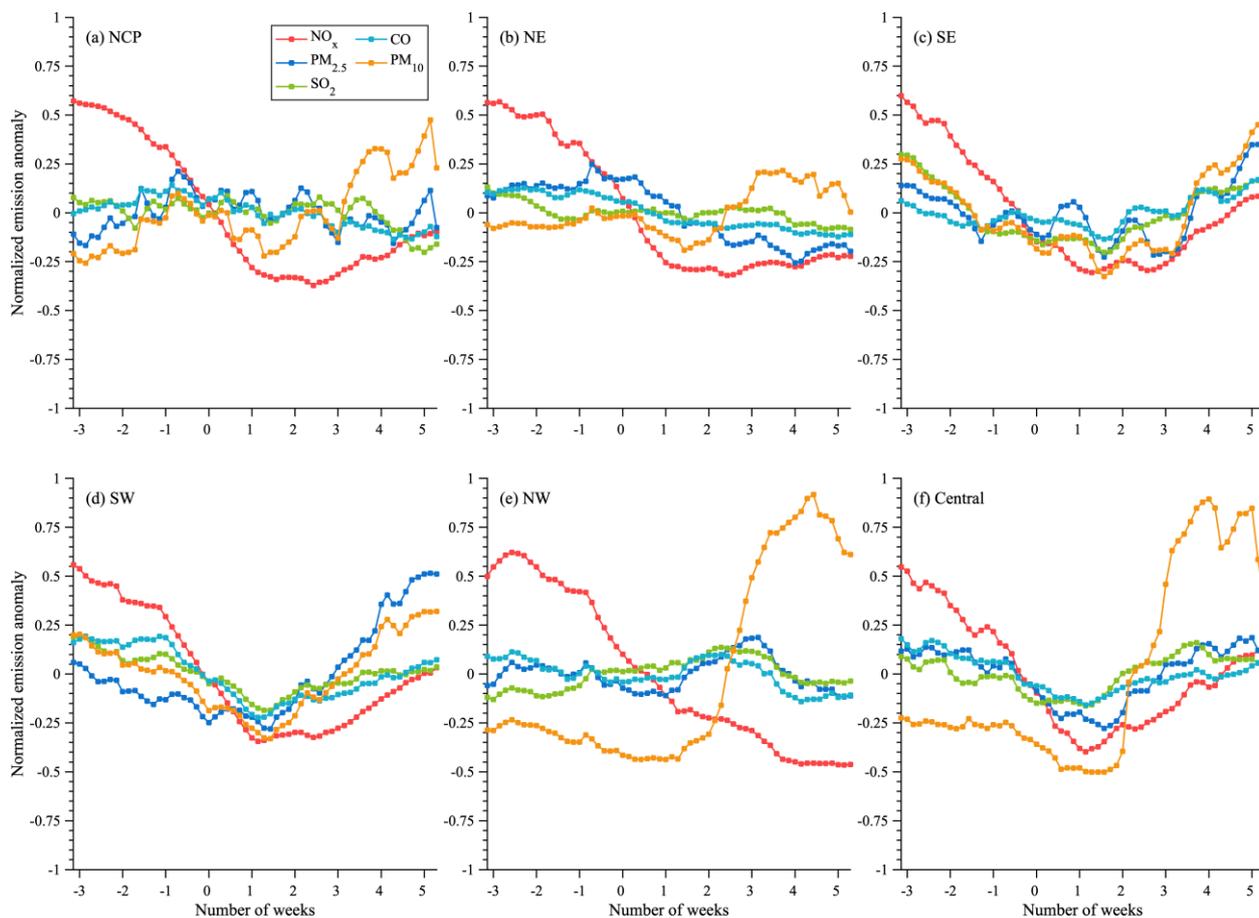
Figure 5: Time series of normalized emission anomalies estimated by inversion results for different species over (a) NCP region, (b) NE region, (c) SE region, (d) SW region, (e) NW region and (f) Central region from 1st January to 29th February 2020.
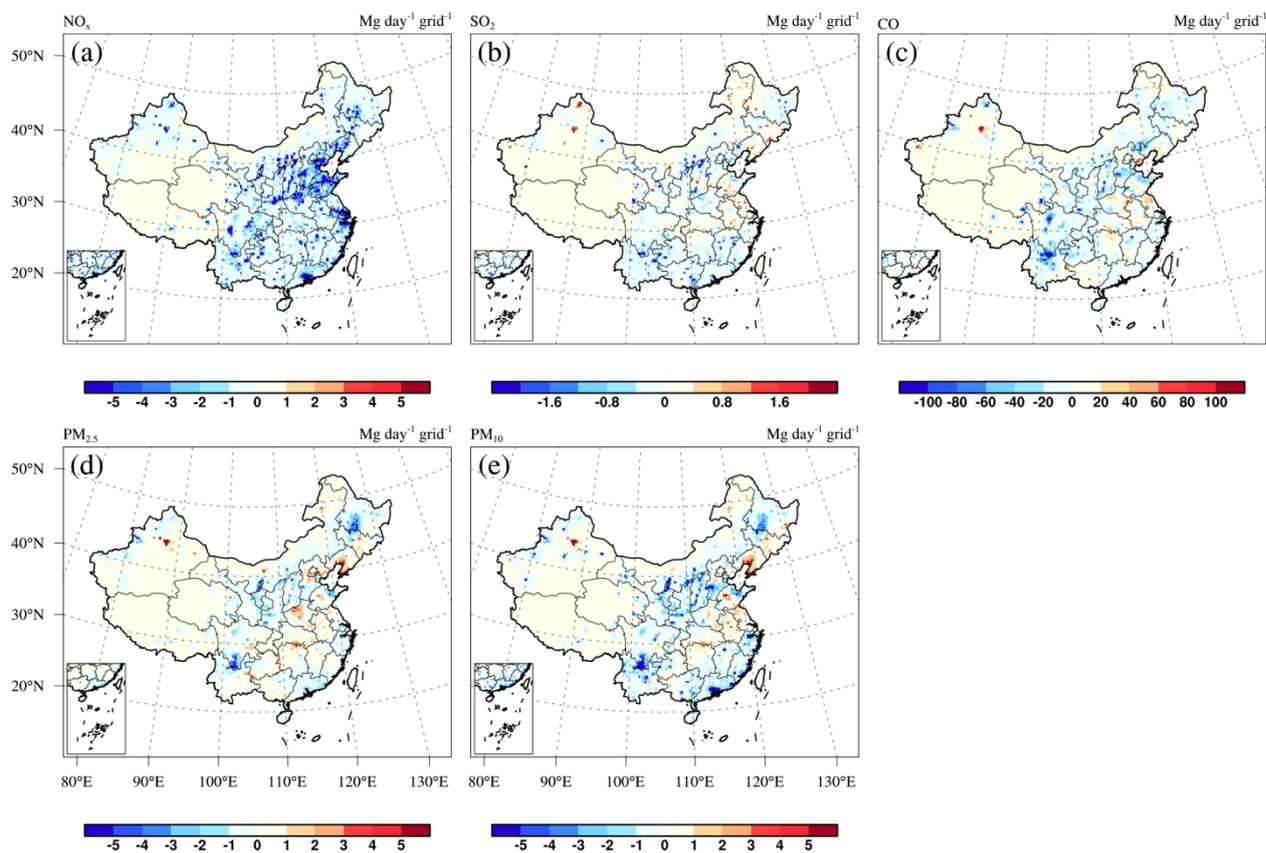
Figure 6: The inversion estimated emission changes of (a) NO$_x$, (b) SO$_2$, (c) CO, (d) PM$_{2.5}$ and (e) PM$_{10}$ in China from P1 to P2 period.
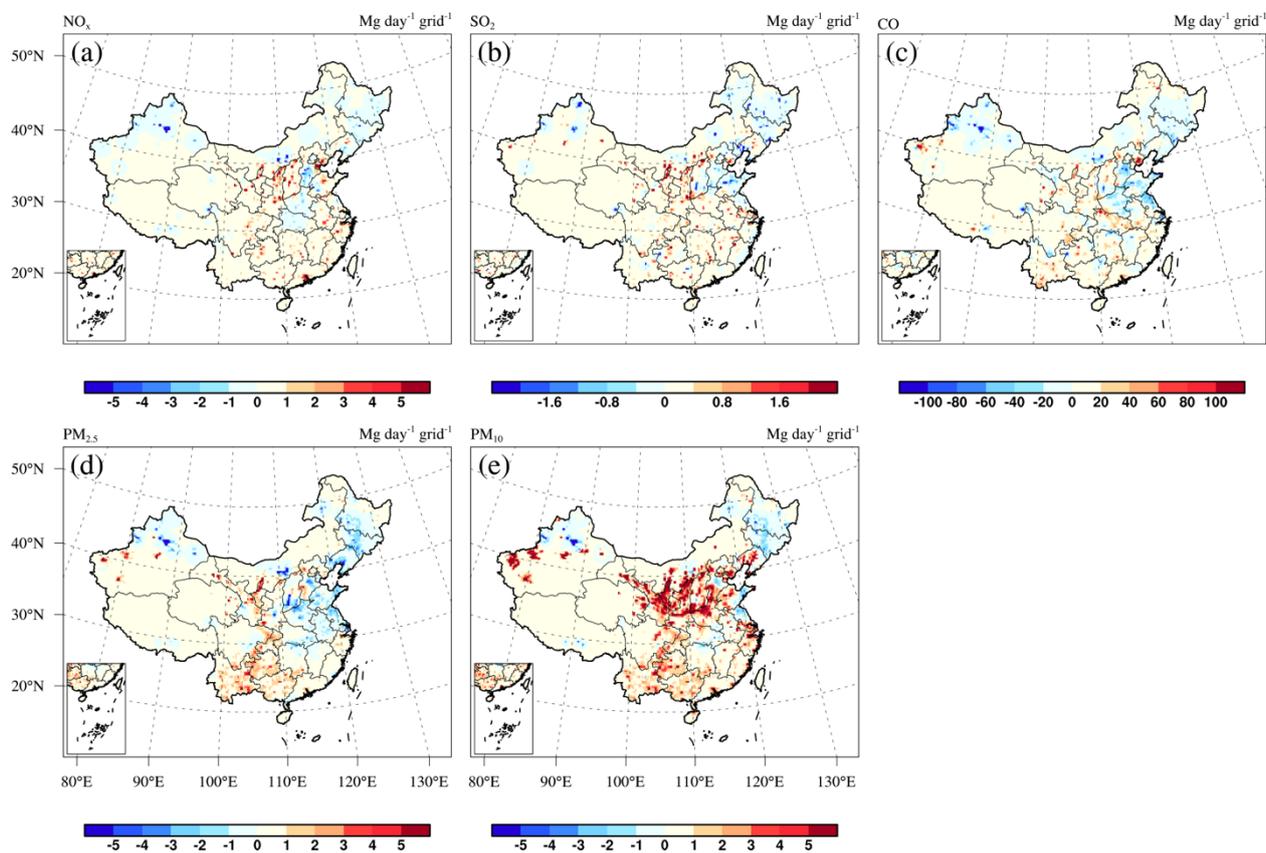
Figure 7: The inversion estimated emission changes of (a) NO$_x$, (b) SO$_2$, (c) CO, (d) PM$_{2.5}$ and (e) PM$_{10}$ in China from P2 to P3 period.
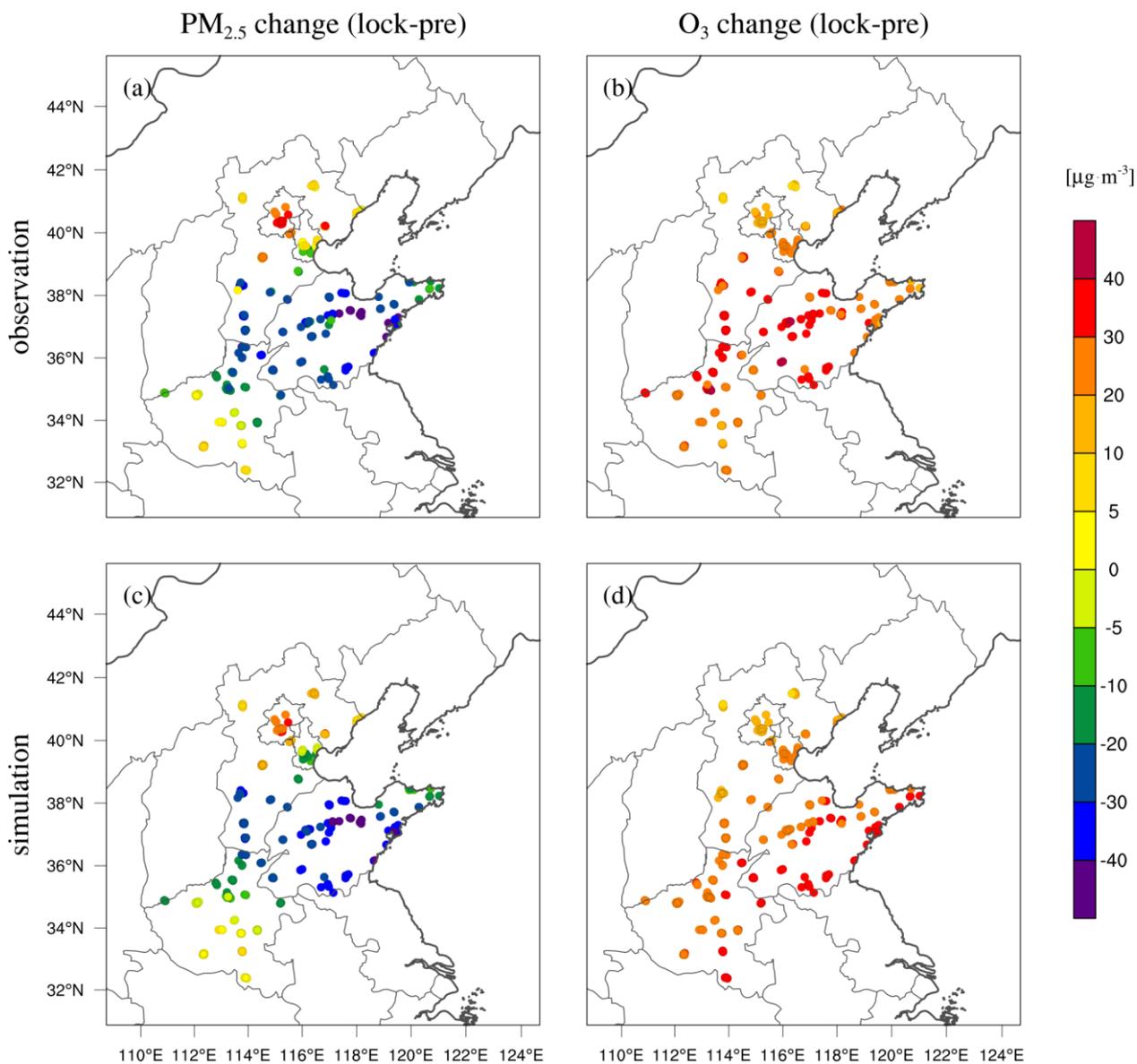
Figure 8: Changes in the observed and simulated concentrations of (a, c) PM₂.₅ and (b, d) O₃ over the NCP region from the pre lockdown period (P1) to the lockdown period (P2).

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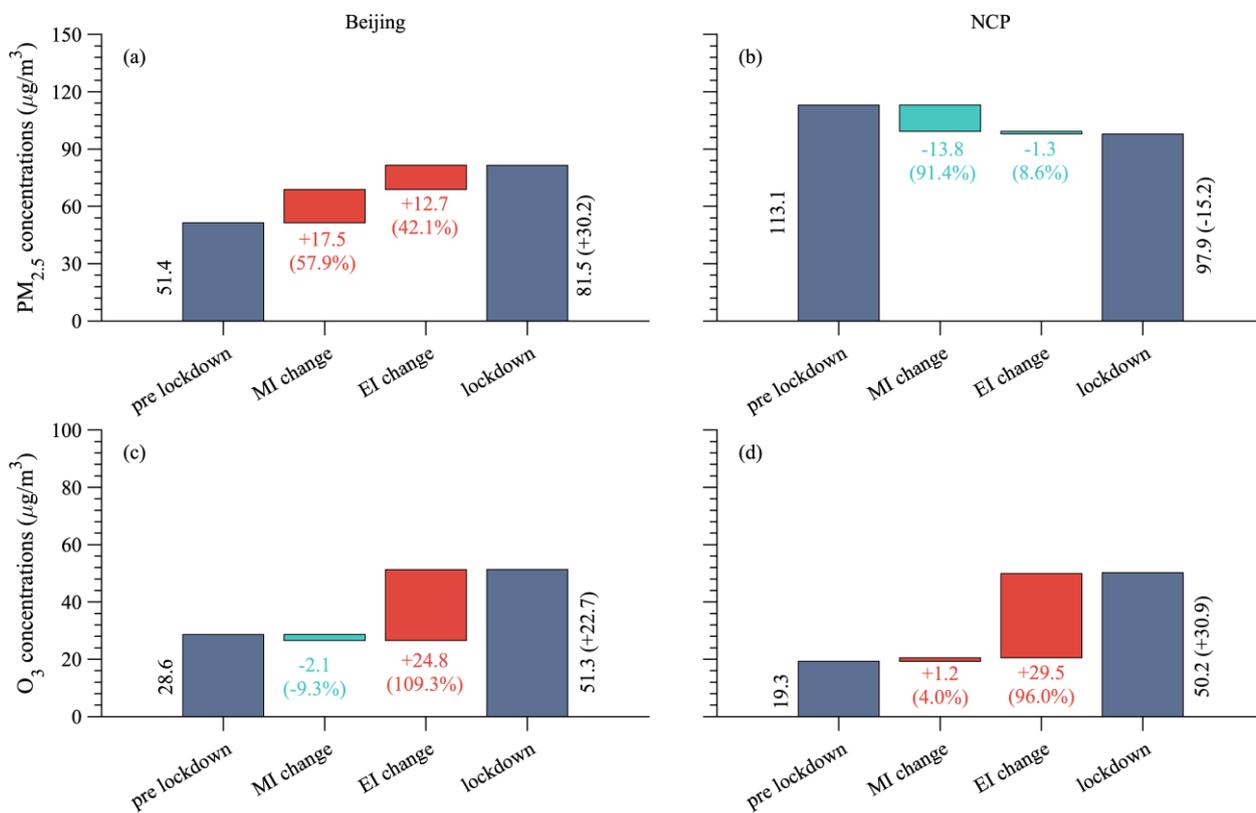
Figure 9: Contributions of the meteorological variations and emission changes to the changes in (a, b) PM$_{2.5}$ and (c, d) O$_3$ concentrations over Beijing and the NCP region from the pre lockdown period (P1) to the lockdown period (P2).
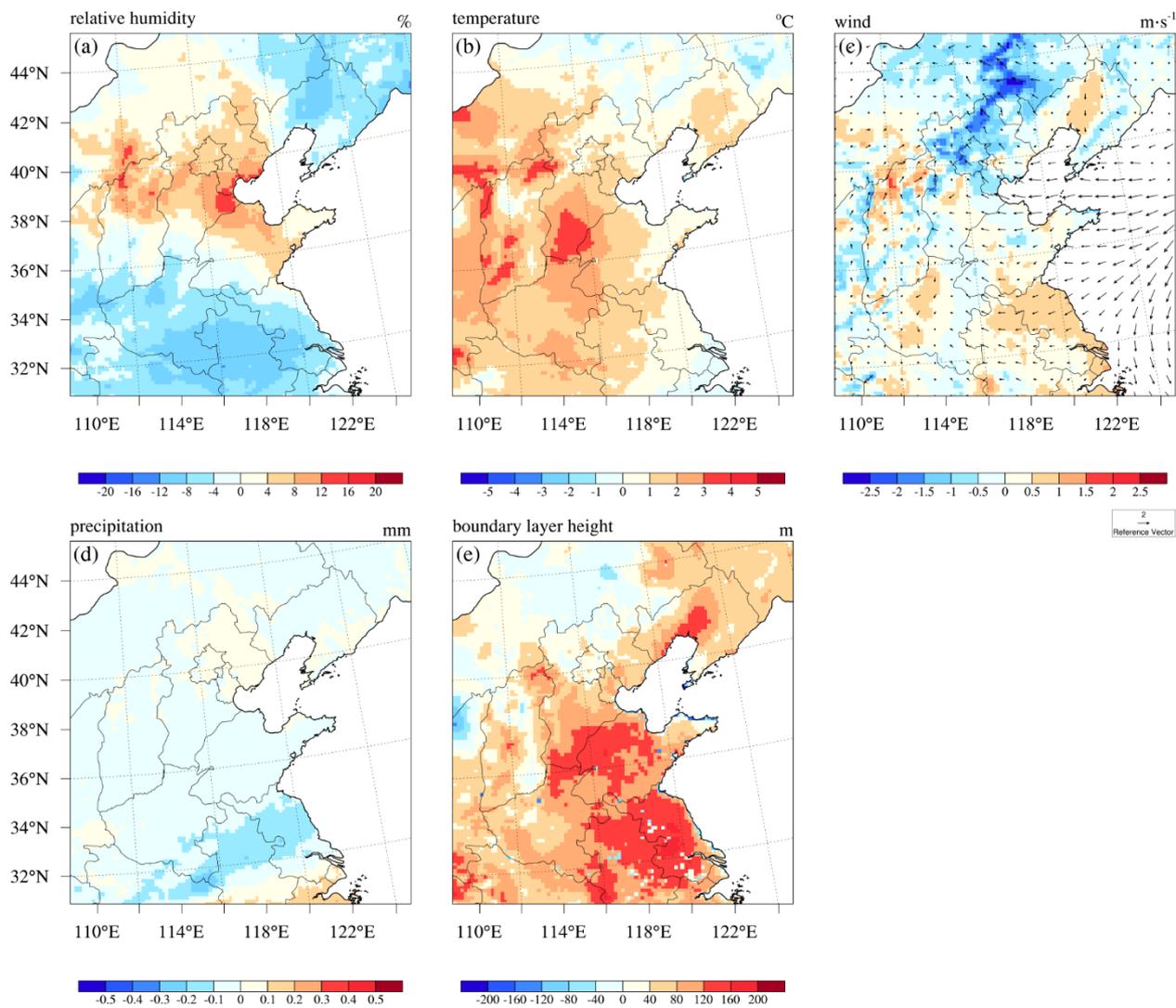
Figure 10: Changes in the (a) relative humidity, (b) temperature, (c) wind speed, (d) precipitation and (e) boundary layer height over the NCP region from P1 to P2 period obtained from WRF simulations.

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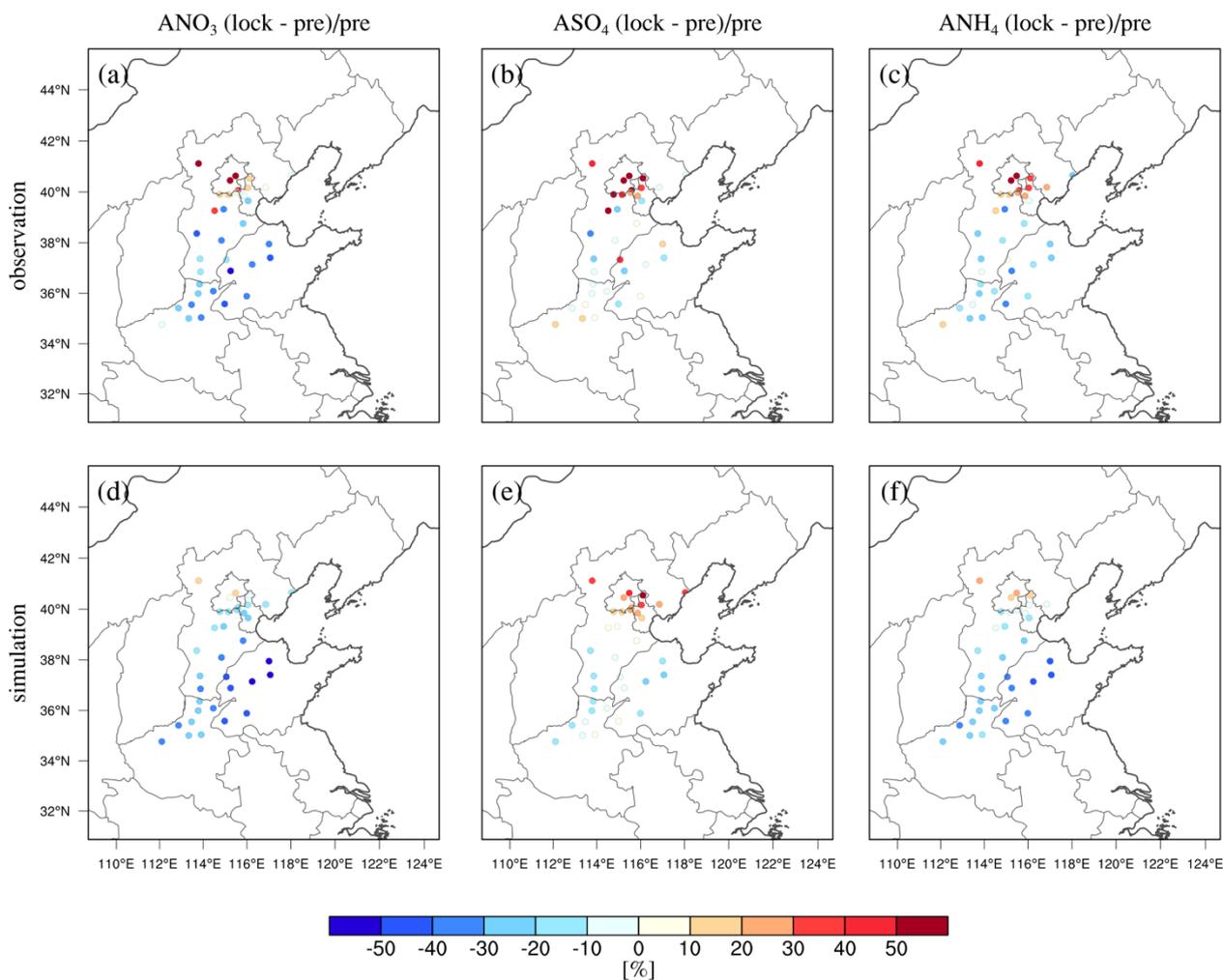
Figure 11: Relative changes in the simulated and observed concentrations of (a) ANO$_3$, (b) ASO$_4$, (c) ANH$_4$ over NCP region from P1 to P2 period.

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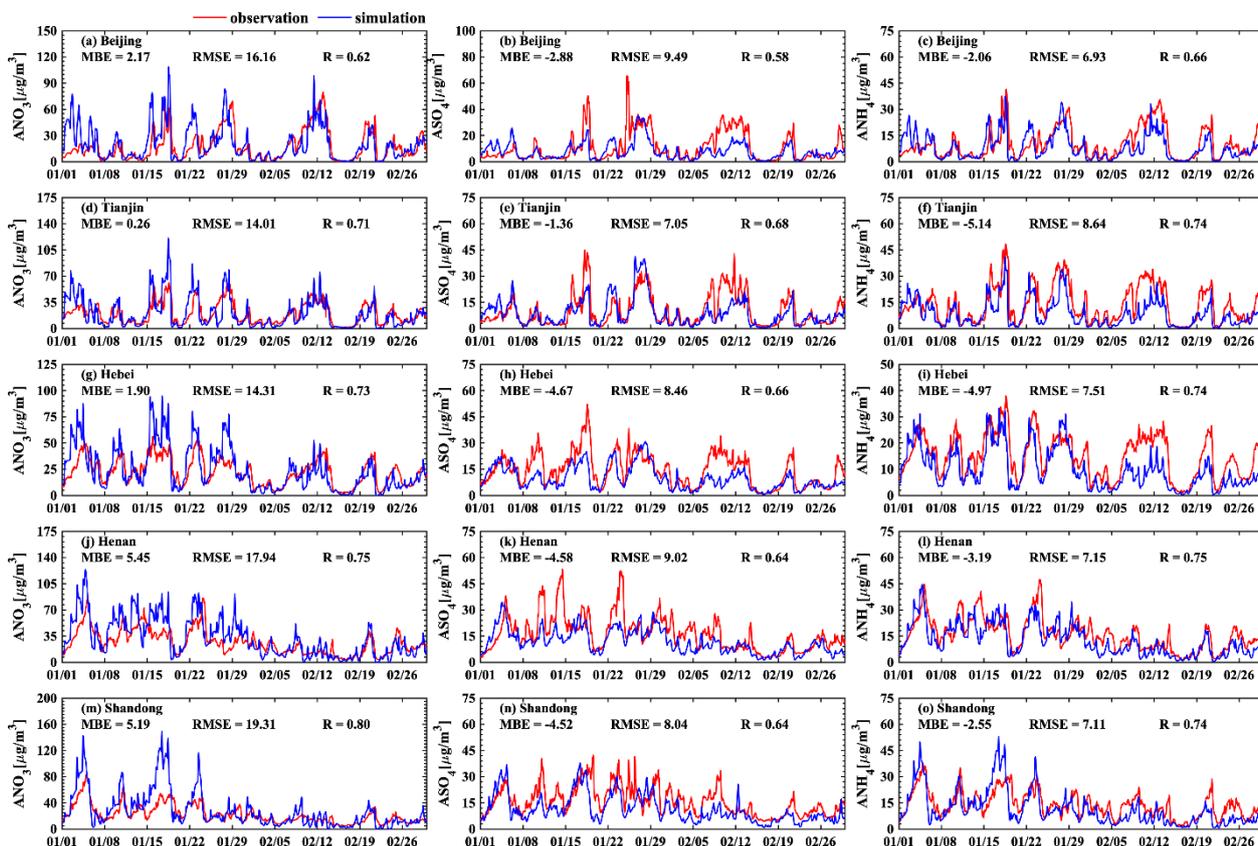
Figure 12: Time series of observed and simulated concentrations of ANO$_3$, ASO$_4$ and ANH$_4$ in (a-c) Beijing, (b-f) Tianjin, (g-i) Heibei, (j-l) Henan and (m-o) Shangdong province from 1st January to 29th February 2020.

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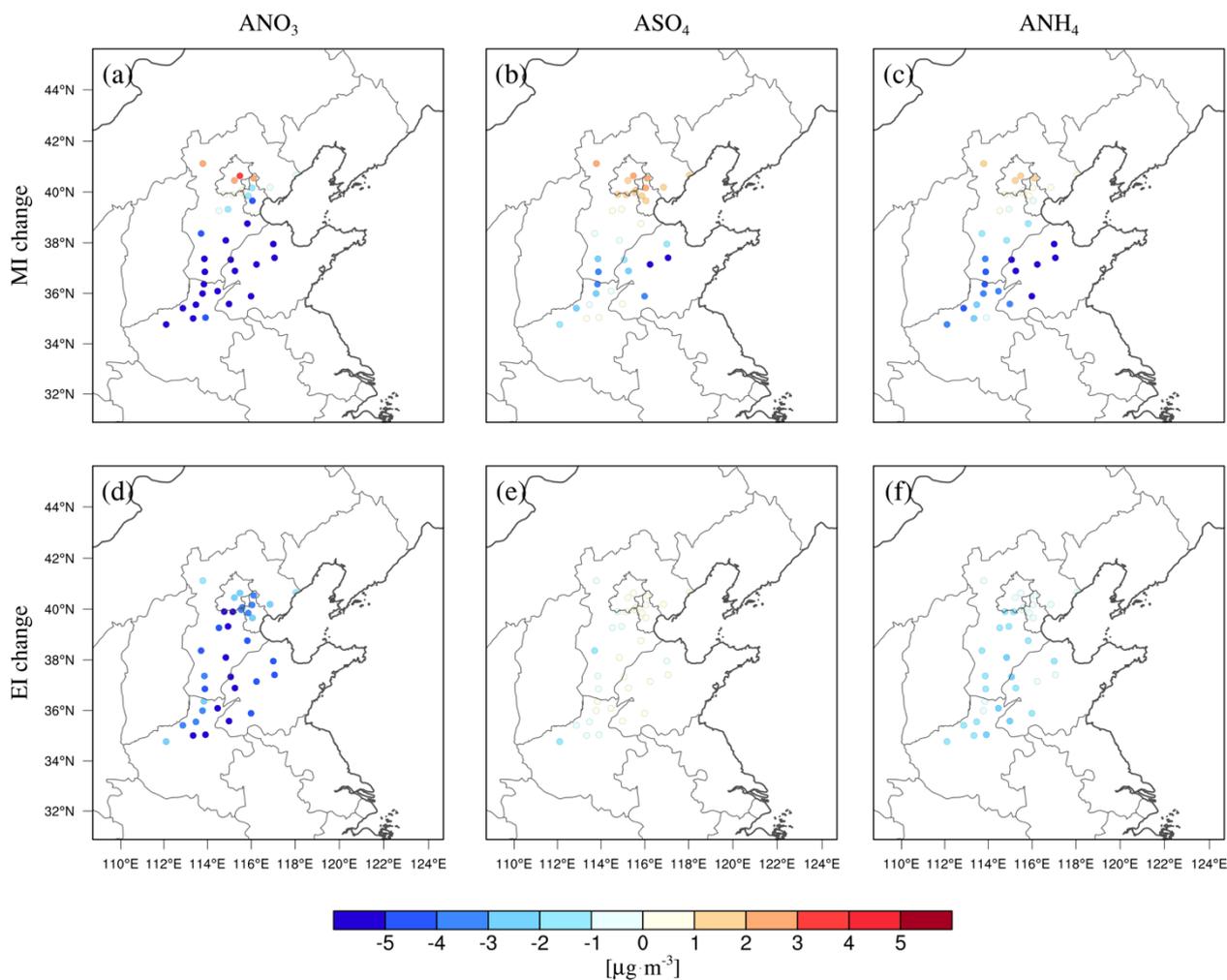
Figure 13: Meteorology-induced (MI) changes in the concentrations of (a) ANO₃, (b) ASO₄ and (c) ANH₄, as well as Emission-induced (EI) changes in the concentrations of (d) ANO₃, (e) ASO₄ and (f) ANH₄.

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Data availability

The hourly surface observations can be obtained from China National Environmental Monitoring Centre (http://www/cnemc.cn/en); The inversion estimated emissions of multi-air pollutants in China during COVID-19 lockdown period and the NAQPMS simulation results are available from the corresponding authors on request.

Author contributions

X.T., J.Z., and Z.W. conceived and designed the project; H.W., L.K., X.T., and L.W. established the data assimilation system; M.L. Q.W. S.H. W.S. contributed to interpreting the data. L.K. conducted the inversion estimate, drew figures, and wrote the paper with comments provided by J.L., X.P., M.G., P.F., Y.S., H.A. and G.R.C.

Competing interests

The authors declare no competing financial interest.

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References

Brasseur, G. P., Hauglustaine, D. A., Walters, S., Rasch, P. J., Muller, J. F., Granier, C., and Tie, X. X.: MOZART, a global chemical transport model for ozone and related chemical tracers 1. Model description, J. Geophys. Res.-Atmos., 103, 28265-28289, https://doi.org/10.1029/98jd02397, 1998.

Cai, H. and Xie, S.: Traffic-related air pollution modeling during the 2008 Beijing Olympic Games: The effects of an odd-even day traffic restriction scheme, Sci. Total Environ., 409, 1935-1948, https://doi.org/10.1016/j.scitotenv.2011.01.025, 2011.

Chen, Z., Hao, X., Zhang, X., and Chen, F.: Have traffic restrictions improved air quality? A shock from COVID-19, J. Clean Prod., 279, 123622, https://doi.org/10.1016/j.jclepro.2020.123622, 2021.

Cheng, C., Barceló, J., Hartnett, A. S., Kubinec, R., and Messerschmidt, L.: COVID-19 Government Response Event Dataset (CoronaNet v.1.0), Nat. Hum. Behav., 4, 756-768, https://doi.org/10.1038/s41562-020-0909-7, 2020.

Chowdhury, S., Dey, S., Tripathi, S. N., Beig, G., Mishra, A. K., and Sharma, S.: "Traffic intervention" policy fails to mitigate air pollution in megacity Delhi, Environ. Sci. Policy, 74, 8-13, https://doi.org/10.1016/j.envsci.2017.04.018, 2017.

Chu, B., Zhang, S., Liu, J., Ma, Q., and He, H.: Significant concurrent decrease in PM2.5 and NO2 concentrations in China during COVID-19 epidemic, J. Environ. Sci., 99, 346-353, https://doi.org/10.1016/j.jes.2020.06.031, 2021.

Chu, K. K., Peng, Z., Liu, Z. Q., Lei, L. L., Kou, X. X., Zhang, Y., Bo, X., and Tian, J.: Evaluating the Impact of Emissions Regulations on the Emissions Reduction During the 2015 China Victory Day Parade With an Ensemble Square Root Filter, J. Geophys. Res.-Atmos., 123, 4122-4134, https://doi.org/10.1002/2017jd027631, 2018.

518 Dai, Q., Liu, B., Bi, X., Wu, J., Liang, D., Zhang, Y., Feng, Y., and Hopke, P. K.: Dispersion Normalized PMF Provides Insights into the
519   Significant Changes in Source Contributions to PM2.5 after the COVID-19 Outbreak, Environ. Sci. Technol., 54, 9917-9927,
520   https://doi.org/10.1021/acs.est.0c02776, 2020.

521 Diamond, M. S. and Wood, R.: Limited Regional Aerosol and Cloud Microphysical Changes Despite Unprecedented Decline in Nitrogen
522   Oxide Pollution During the February 2020 COVID-19 Shutdown in China, Geophys. Res. Lett., 47, e2020GL088913,
523   https://doi.org/10.1029/2020gl088913, 2020.

524 Evensen, G.: Sequential data assimilation with a nonlinear quasi-geostrophic model using Monte Carlo methods to forecast error statistics,
525   J. Geophys. Res.-Oceans, 99, 10143-10162, https://doi.org/10.1029/94JC00572, 1994.

526 Fan, C., Li, Y., Guang, J., Li, Z. Q., Elnashar, A., Allam, M., and de Leeuw, G.: The Impact of the Control Measures during the COVID-19
527   Outbreak on Air Pollution in China, Remote Sens., 12, 23, https://doi.org/10.3390/rs12101613, 2020.

528 Feng, S., Jiang, F., Wang, H., Wang, H., Ju, W., Shen, Y., Zheng, Y., Wu, Z., and Ding, A.: NOx Emission Changes Over China During the
529   COVID-19 Epidemic Inferred From Surface NO2 Observations, Geophys. Res. Lett., 47, e2020GL090080, 10.1029/2020gl090080, 2020.

530 Forster, P. M., Forster, H. I., Evans, M. J., Gidden, M. J., Jones, C. D., Keller, C. A., Lamboll, R. D., Le Quere, C., Rogelj, J., Rosen, D.,
531   Schleussner, C. F., Richardson, T. B., Smith, C. J., and Turnock, S. T.: Current and future global climate impacts resulting from COVID-
532   19, Nat. Clim. Chang., 10, 913-+, https://doi.org/10.1038/s41558-020-0883-0, 2020.

533 Granier, C., Lamarque, J., Mieville, A., Muller, J., Olivier, J., Orlando, J., Peters, J., Petron, G., Tyndall, G., and Wallens, S.: POET, a
534   database of surface emissions of ozone precursors, 2005.

535 Hammer, M. S., van Donkelaar, A., Martin, R. V., McDuffie, E. E., Lyapustin, A., Sayer, A. M., Hsu, N. C., Levy, R. C., Garay, M. J.,
536   Kalashnikova, O. V., and Kahn, R. A.: Effects of COVID-19 lockdowns on fine particulate matter concentrations, Sci. Adv., 7,
537   https://doi.org/10.1126/sciadv.abg7670, 2021.

538 Han, X. L. and Naeher, L. P.: A review of traffic-related air pollution exposure assessment studies in the developing world, Environ. Int., 32,
539   106-120, https://doi.org/10.1016/j.envint.2005.05.020, 2006.

540 Hauglustaine, D. A., Brasseur, G. P., Walters, S., Rasch, P. J., Muller, J. F., Emmons, L. K., and Carroll, C. A.: MOZART, a global
541   chemical transport model for ozone and related chemical tracers 2. Model results and evaluation, J. Geophys. Res.-Atmos., 103, 28291-
542   28335, https://doi.org/10.1029/98jd02398, 1998.

543 He, G. J., Pan, Y. H., and Tanaka, T.: The short-term impacts of COVID-19 lockdown on urban air pollution in China, Nat. Sustain, 3, 9,
544   https://doi.org/10.1038/s41893-020-0581-y, 2020.

545 Hu, Y., Zang, Z., Ma, X., Li, Y., Liang, Y., You, W., Pan, X., and Li, Z.: Four-dimensional variational assimilation for SO2 emission and
546   its application around the COVID-19 lockdown in the spring 2020 over China, Atmos. Chem. Phys., 22, 13183-13200,
547   https://doi.org/10.5194/acp-22-13183-2022, 2022.

548 Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., Chi, X., Xu, Z., Chen, L., Li, Y., Che, F.,
549   Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu, W., Fu, Q., Liu, B., Chai, F., Davis, S. J., Zhang, Q., and He, K.: Enhanced
550   secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China, Natl. Sci. Rev., 8,
551   https://doi.org/10.1093/nsr/nwaa137, 2021.

552 Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J.,
553   Wankmuller, R., van der Gon, H. D., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP_v2.2: a mosaic of
554   regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, Atmos. Chem. Phys., 15,
555   11411-11432, https://doi.org/10.5194/acp-15-11411-2015, 2015.

556 Kong, L., Tang, X., Zhu, J., Wang, Z., Pan, Y., Wu, H., Wu, L., Wu, Q., He, Y., Tian, S., Xie, Y., Liu, Z., Sui, W., Han, L., and Carmichael,
557   G.: Improved Inversion of Monthly Ammonia Emissions in China Based on the Chinese Ammonia Monitoring Network and Ensemble
558   Kalman Filter, Environ. Sci. Technol., 53, 12529-12538, https://doi.org/10.1021/acs.est.9b02701, 2019.

559 Kong, L., Tang, X., Zhu, J., Wang, Z., Li, J., Wu, H., Wu, Q., Chen, H., Zhu, L., Wang, W., Liu, B., Wang, Q., Chen, D., Pan, Y., Song, T.,
560   Li, F., Zheng, H., Jia, G., Lu, M., Wu, L., and Carmichael, G. R.: A 6-year-long (2013–2018) high-resolution air quality reanalysis
561   dataset in China based on the assimilation of surface observations from CNEMC, Earth Syst. Sci. Data, 13, 529-570,
562   https://doi.org/10.5194/essd-13-529-2021, 2021.

563 Le, T. H., Wang, Y., Liu, L., Yang, J. N., Yung, Y. L., Li, G. H., and Seinfeld, J. H.: Unexpected air pollution with marked emission
564   reductions during the COVID-19 outbreak in China, Science, 369, 702-+, https://doi.org/10.1126/science.abb7431, 2020.

565 Levelt, P. F., Stein Zweers, D. C., Aben, I., Bauwens, M., Borsdorff, T., De Smedt, I., Eskes, H. J., Lerot, C., Loyola, D. G., Romahn, F.,
566   Stavrakou, T., Theys, N., Van Roozendael, M., Veefkind, J. P., and Verhoelst, T.: Air quality impacts of COVID-19 lockdown measures
567   detected from space using high spatial resolution observations of multiple trace gases from Sentinel-5P/TROPOMI, Atmos. Chem. Phys.,
568   22, https://doi.org/10319-10351, 10.5194/acp-22-10319-2022, 2022.

569 Li, F., Tang, X., Wang, Z., Zhu, L., Wang, X., Wu, H., Lu, M., Li, J., and Zhu, J.: Estimation of Representative Errors of Surface
570   Observations of Air Pollutant Concentrations Based on High-Density Observation Network over Beijing-Tianjin-Hebei Region, Chinese
571   Journal of Atmospheric Sciences, 43, 277-284, 2019.

572 Li, L., Li, Q., Huang, L., Wang, Q., Zhu, A., Xu, J., Liu, Z., Li, H., Shi, L., Li, R., Azari, M., Wang, Y., Zhang, X., Liu, Z., Zhu, Y., Zhang,
573   K., Xue, S., Ooi, M. C. G., Zhang, D., and Chan, A.: Air quality changes during the COVID-19 lockdown over the Yangtze River Delta
574   Region: An insight into the impact of human activity pattern changes on air pollution variation, Sci. Total Environ., 732, 139282,
575   https://doi.org/10.1016/j.scitotenv.2020.139282, 2020.

576 Li, M., Wang, T., Xie, M., Li, S., Zhuang, B., Fu, Q., Zhao, M., Wu, H., Liu, J., Saikawa, E., and Liao, K.: Drivers for the poor air quality
577   conditions   in   North   China   Plain   during   the   COVID-19   outbreak,   Atmos.   Environ.,   246,   118103,
578   https://doi.org/10.1016/j.atmosenv.2020.118103, 2021.

579 Li, M., Liu, H., Geng, G. N., Hong, C. P., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H. Y., Man, H. Y., Zhang, Q., and He, K. B.:
580   Anthropogenic emission inventories in China: a review, Natl. Sci. Rev., 4, 834-866, https://doi.org/10.1093/nsr/nwx150, 2017a.

581 Li, M., Zhang, Q., Kurokawa, J. I., Woo, J. H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C.,
582   Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the
583   international collaboration framework of the MICS-Asia and HTAP, Atmos. Chem. Phys., 17, 935-963, https://doi.org/10.5194/acp-17-
584   935-2017, 2017b.

585 Li, X., Zhang, Q., Zhang, Y., Zhang, L., Wang, Y. X., Zhang, Q. Q., Li, M., Zheng, Y. X., Geng, G. N., Wallington, T. J., Han, W. J., Shen,
586   W., and He, K. B.: Attribution of PM2.5 exposure in Beijing-Tianjin-Hebei region to emissions: implication to control strategies, Sci.
587   Bull., 62, 957-964, https://doi.org/10.1016/j.scib.2017.06.005, 2017c.

588 Ma, C. Q., Wang, T. J., Mizzi, A. P., Anderson, J. L., Zhuang, B. L., Xie, M., and Wu, R. S.: Multiconstituent Data Assimilation With
589   WRF-Chem/DART: Potential for Adjusting Anthropogenic Emissions and Improving Air Quality Forecasts Over Eastern China, J.
590   Geophys. Res.-Atmos., 124, 7393-7412, https://doi.org/10.1029/2019jd030421, 2019.

591 Miyazaki, K., Eskes, H. J., Sudo, K., Takigawa, M., van Weele, M., and Boersma, K. F.: Simultaneous assimilation of satellite NO2, O-3,
592   CO, and HNO3 data for the analysis of tropospheric chemical composition and emissions, Atmos. Chem. Phys., 12, 9545-9579,
593   https://doi.org/10.5194/acp-12-9545-2012, 2012.

594 Okuda, T., Matsuura, S., Yamaguchi, D., Umemura, T., Hanada, E., Orihara, H., Tanaka, S., He, K., Ma, Y., Cheng, Y., and Liang, L.: The

595   impact of the pollution control measures for the 2008 Beijing Olympic Games on the chemical composition of aerosols, Atmos. Environ.,

596   45, 2789-2794, https://doi.org/10.1016/j.atmosenv.2011.01.053, 2011.

597 Peng, Z., Lei, L. L., Liu, Z. Q., Su, J. N., Ding, A. J., Ban, J. M., Chen, D., Kou, X. X., and Chu, K. K.: The impact of multi-species surface

598   chemical observation assimilation on air quality forecasts in China, Atmos. Chem. Phys., 18, 18, https://doi.org/10.5194/acp-18-17387-

599   2018, 2018.

600 Price, C., Penner, J., and Prather, M.: NOx from lightning .1. Global distribution based on lightning physics, J. Geophys. Res.-Atmos., 102,

601   5929-5941, https://doi.org/10.1029/96jd03504, 1997.

602 Randerson, J. T., Van Der Werf, G. R., Giglio, L., Collatz, G. J., and Kasibhatla, P. S.: Global Fire Emissions Database, Version 4.1

603   (GFEDv4), 10.3334/ornldaac/1293, 2017.

604 Sakov, P. and Oke, P. R.: A deterministic formulation of the ensemble Kalman filter: an alternative to ensemble square root filters, Tellus

605   Ser. A-Dyn. Meteorol. Oceanol., 60, 361-371, https://doi.org/10.1111/j.1600-0870.2007.00299.x, 2008.

606 Shi, X. and Brasseur, G. P.: The Response in Air Quality to the Reduction of Chinese Economic Activities During the COVID-19 Outbreak,

607   Geophys. Res. Lett., 47, e2020GL088070, https://doi.org/10.1029/2020gl088070, 2020.

608 Shi, Z., Song, C., Liu, B., Lu, G., Xu, J., Van Vu, T., Elliott, R. J. R., Li, W., Bloss, W. J., and Harrison, R. M.: Abrupt but smaller than

609   expected changes in surface air quality attributable to COVID-19 lockdowns, Sci. Adv., 7, eabd6696,

610   https://doi.org/10.1126/sciadv.abd6696, 2021.

611 Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Muller, J. F., Kuhn, U., Stefani, P., and Knorr, W.: Global

612   data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, Atmos. Chem. Phys., 14, 9317-9341,

613   https://doi.org/10.5194/acp-14-9317-2014, 2014.

614 Skamarock, W. C.: A description of the advanced research WRF version 3, Ncar Technical, 113, 7-25, 2008.

615 Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo,

616   J. H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res.-Atmos., 108,

617   23, https://doi.org/10.1029/2002JD003093, 2003.

618 Sulaymon, I. D., Zhang, Y., Hopke, P. K., Hu, J., Zhang, Y., Li, L., Mei, X., Gong, K., Shi, Z., Zhao, B., and Zhao, F.: Persistent high

619   PM2.5 pollution driven by unfavorable meteorological conditions during the COVID-19 lockdown period in the Beijing-Tianjin-Hebei

620   region, China, Environ. Res., 198, 111186, https://doi.org/10.1016/j.envres.2021.111186, 2021.

621 Tang, G., Zhu, X., Hu, B., Xin, J., Wang, L., Münkel, C., Mao, G., and Wang, Y.: Impact of emission controls on air quality in Beijing

622   during APEC 2014: lidar ceilometer observations, Atmos. Chem. Phys., 15, 12667-12680, https://doi.org/10.5194/acp-15-12667-2015,

623   2015.

624 Tang, X., Zhu, J., Wang, Z. F., and Gbaguidi, A.: Improvement of ozone forecast over Beijing based on ensemble Kalman filter with

625   simultaneous adjustment of initial conditions and emissions, Atmos. Chem. Phys., 11, 12901-12916, 10.5194/acp-11-12901-2011, 2011.

626 Tang, X., Zhu, J., Wang, Z. F., Wang, M., Gbaguidi, A., Li, J., Shao, M., Tang, G. Q., and Ji, D. S.: Inversion of CO emissions over Beijing

627   and its surrounding areas with ensemble Kalman filter, Atmos. Environ., 81, 676-686, https://doi.org/10.1016/j.atmosenv.2013.08.051,

628   2013.

629 van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van

630   Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009),

631   Atmos. Chem. Phys., 10, 11707-11735, https://doi.org/10.5194/acp-10-11707-2010, 2010.

632 Wang, P., Chen, K., Zhu, S., Wang, P., and Zhang, H.: Severe air pollution events not avoided by reduced anthropogenic activities during
633   COVID-19 outbreak, Resour. Conserv. Recycl., 158, 104814, https://doi.org/10.1016/j.resconrec.2020.104814, 2020.

634 Wang, S., Gao, J., Zhang, Y., Zhang, J., Cha, F., Wang, T., Ren, C., and Wang, W.: Impact of emission control on regional air quality: An
635   observational study of air pollutants before, during and after the Beijing Olympic Games, J. Environ. Sci., 26, 175-180,
636   https://doi.org/10.1016/S1001-0742(13)60395-2, 2014.

637 Wang, Z., Uno, I., Yumimoto, K., Itahashi, S., Chen, X., Yang, W., and Wang, Z.: Impacts of COVID-19 lockdown, Spring Festival and
638   meteorology on the NO2 variations in early 2020 over China based on in-situ observations, satellite retrievals and model simulations,
639   Atmos. Environ., 244, 117972, https://doi.org/10.1016/j.atmosenv.2020.117972, 2021.

640 Wu, H., Tang, X., Wang, Z., Wu, L., Li, J., Wang, W., Yang, W., and Zhu, J.: High-spatiotemporal-resolution inverse estimation of CO and
641   NOx emission reductions during emission control periods with a modified ensemble Kalman filter, Atmos. Environ., 236, 117631,
642   https://doi.org/10.1016/j.atmosenv.2020.117631, 2020.

643 Wu, H. J., Tang, X., Wang, Z. F., Wu, L., Lu, M. M., Wei, L. F., and Zhu, J.: Probabilistic Automatic Outlier Detection for Surface Air
644   Quality Measurements from the China National Environmental Monitoring Network, Adv. Atmos. Sci., 35, 1522-1532,
645   https://doi.org/10.1007/s00376-018-8067-9, 2018.

646 Xing, J., Li, S. W., Jiang, Y. Q., Wang, S. X., Ding, D., Dong, Z. X., Zhu, Y., and Hao, J. M.: Quantifying the emission changes and
647   associated air quality impacts during the COVID-19 pandemic on the North China Plain: a response modeling study, Atmos. Chem.
648   Phys., 20, 14347-14359, https://doi.org/10.5194/acp-20-14347-2020, 2020.

649 Yan, C., Shen, Y. C., Stolzenburg, D., Dada, L., Qi, X. M., Hakala, S., Sundstrom, A. M., Guo, Y. S., Lipponen, A., Kokkonen, T. V.,
650   Kontkanen, J., Cai, R. L., Cai, J., Chan, T., Chen, L. D., Chu, B. W., Deng, C. J., Du, W., Fan, X. L., He, X. C., Kangasluoma, J.,
651   Kujansuu, J., Kurppa, M., Li, C., Li, Y. R., Lin, Z. H., Liu, Y. L., Liu, Y. L., Lu, Y. Q., Nie, W., Pulliainen, J., Qiao, X. H., Wang, Y. H.,
652   Wen, Y. F., Wu, Y., Yang, G., Yao, L., Yin, R. J., Zhang, G., Zhang, S. J., Zheng, F. X., Zhou, Y., Arola, A., Tamminen, J., Paasonen,
653   P., Sun, Y. L., Wang, L., Donahue, N. M., Liu, Y. C., Bianchi, F., Daellenbach, K. R., Worsnop, D. R., Kerminen, V. M., Petaja, T.,
654   Ding, A. J., Jiang, J. K., and Kulmala, M.: The effect of COVID-19 restrictions on atmospheric new particle formation in Beijing, Atmos.
655   Chem. Phys., 22, 12207-12220, https://doi.org/10.5194/acp-22-12207-2022, 2022.

656 Yan, X. Y., Akimoto, H., and Ohara, T.: Estimation of nitrous oxide, nitric oxide and ammonia emissions from croplands in East, Southeast
657   and South Asia, Glob. Change Biol., 9, 1080-1096, https://doi.org/10.1046/j.1365-2486.2003.00649.x, 2003.

658 Zhang, L., Shao, J., Lu, X., Zhao, Y., Hu, Y., Henze, D. K., Liao, H., Gong, S., and Zhang, Q.: Sources and Processes Affecting Fine
659   Particulate Matter Pollution over North China: An Adjoint Analysis of the Beijing APEC Period, Environ. Sci. Technol., 50, 8731-8740,
660   https://doi.org/10.1021/acs.est.6b03010, 2016.

661 Zhang, Q., Pan, Y., He, Y., Walters, W. W., Ni, Q., Liu, X., Xu, G., Shao, J., and Jiang, C.: Substantial nitrogen oxides emission reduction
662   from China due to COVID-19 and its impact on surface ozone and aerosol pollution, Sci. Total Environ., 753, 142238,
663   https://doi.org/10.1016/j.scitotenv.2020.142238, 2021.

664 Zhang, R. X., Zhang, Y. Z., Lin, H. P., Feng, X., Fu, T. M., and Wang, Y. H.: NOx Emission Reduction and Recovery during COVID-19 in
665   East China, Atmosphere, 11, 15, https://doi.org/10.3390/atmos11040433, 2020.

666 Zhang, W., Guo, J. H., Sun, Y. L., Yuan, H., Zhuang, G. S., Zhuang, Y. H., and Hao, Z. P.: Source apportionment for,urban PM10 and
667   PM2.5 in the Beijing area, Chin. Sci. Bull., 52, 608-615, https://doi.org/10.1007/s11434-007-0076-5, 2007.

668 Zhao, X., Wang, G., Wang, S., Zhao, N., Zhang, M., and Yue, W.: Impacts of COVID-19 on air quality in mid-eastern China: An insight
669   into meteorology and emissions, Atmos. Environ., 266, 118750, https://doi.org/10.1016/j.atmosenv.2021.118750, 2021.

670 Zhao, Y. B., Zhang, K., Xu, X. T., Shen, H. Z., Zhu, X., Zhang, Y. X., Hu, Y. T., and Shen, G. F.: Substantial Changes in Nitrogen Dioxide
671   and Ozone after Excluding Meteorological Impacts during the COVID-19 Outbreak in Mainland China, Environ. Sci. Technol. Lett., 7,
672   402-408, https://doi.org/10.1021/acs.estlett.0c00304, 2020.
673 Zheng, B., Zhang, Q., Geng, G., Chen, C., Shi, Q., Cui, M., Lei, Y., and He, K.: Changes in China's anthropogenic emissions and air quality
674   during the COVID-19 pandemic in 2020, Earth Syst. Sci. Data, 13, 2895-2907, https://doi.org/10.5194/essd-13-2895-2021, 2021.
675 Zheng, B., Tong, D., Li, M., Liu, F., Hong, C. P., Geng, G. N., Li, H. Y., Li, X., Peng, L. Q., Qi, J., Yan, L., Zhang, Y. X., Zhao, H. Y.,
676   Zheng, Y. X., He, K. B., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions,
677   Atmos. Chem. Phys., 18, 14095-14111, https://doi.org/10.5194/acp-18-14095-2018, 2018.
678

679