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 NOₓ, SO₂, CO, PM₂.₅ and PM₁₀ 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 NOₓ 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 PM₂.₅ concentrations occurred over North China Plain (NCP) during lockdown period, the emission change only accounted for 8.6% of PM₂.₅ reductions, and even led to substantial increases of O₃. The meteorological variation instead dominated the changes in PM₂.₅ concentrations over NCP, which contributed 90%
of the PM$_{2.5}$ reductions over most parts of NCP region. Meanwhile, our results also suggest that the local stagnant meteorological conditions together with inefficient reductions in PM$_{2.5}$ emissions were the main drivers of the unexpected COVID-19 haze in Beijing. These results highlighted that traffic control as a separate pollution control measure has limited effects on the coordinated control of O$_3$ and PM$_{2.5}$ concentrations under current complex air pollution conditions in China.

More comprehensive and balanced regulations for multiple precursors from different sectors are required to address O$_3$ and PM$_{2.5}$ pollution in China.

1 Introduction

A novel coronavirus disease (COVID-19) broke out in Wuhan at the end of 2019 but quickly spread across the whole China within a month. To curb the spread of the virus, strict epidemic control measures were implemented by Chinese governments to prevent large gatherings, including strict travel restriction, shutting down of non-essential industries, extended holidays, closing of schools and entertainment houses (Cheng et al., 2020). These restrictions have had a significant impact on the industrial activities and social life, as exemplified by the drop of China’s industrial output by 15-30% (https://data.stats.gov.cn/ last accessed on 22 Oct, 2022) and the dramatic decrease of traffic flow by 60–90% in major cities of China during COVID-19 epidemic (http://jiaotong.baidu.com/, last accessed on 22 Oct, 2022), which provides us a natural experiment to examine the responses of the emissions and air quality on the changes in human activities.

It has been well documented that the short-term stringent emission control targeted on power generator or heavy industry enacted by Chinese government during certain societal events, such as the 2008 Olympics Games, 2014 Asia-Pacific Economic Cooperation conference and 2015 China Victory Day Parade, is an effective way to reduce emissions and 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., 2018). However, different from those stringent emission controls, the COVID-19 restrictions are inclined to affect emissions from sectors more closely to social life whose influence on emissions has still not well been assessed. Previous studies suggest that the COVID-19 restrictions have substantially reduced the China’s anthropogenic emissions from almost all sectors (Zheng et al., 2021; Huang et al., 2021; Xing et al., 2020). For example, by using a bottom-up method based on near-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%, 27%, 28% and 24% during COVID-19 restrictions, mostly due to the reductions in industry and transportation sector. Xing et al. (2020), by using a response model, estimated stronger COVID-19 shutdown effects on emissions over the North China Plain (NCP) with emissions of NO$_x$, SO$_2$ and primary PM$_{2.5}$ dropped by 51%, 28% and 63%, respectively. Others argue that the COVID-19 restriction may mainly affect the emissions from transportation, light industry and manufacturing, while it has much smaller effects on the emissions from the power generator and heavy industry because of their non-interruptible processes (Chu et al., 2021; Hammer et al., 2021; Le et al., 2020; Zhao et al., 2020). Moreover, the residential emissions may even increase during the COVID-19 lockdown due to the increased demanding for space heating and cooking with the stay-at-home orders. Therefore, Le et al. (2020) only considered the NO$_x$ reductions during COVID-19 restrictions in their
investigation of the severe haze during COVID-19 lockdown, and similarly, Hammer et al. (2021) only considered the emission reductions in the transportation sector. This indicates that there has large uncertainty in the current understanding of the effects of COVID-19 restrictions on the emissions of different species.

Quantification of the emission changes of different species and different sectors during the COVID-19 lockdown is thus necessary for the comprehensive understanding of the environmental impacts of COVID-19 restrictions. In particular, although observations indeed show decreases of air pollutant concentrations during COVID-19 restrictions (Fan et al., 2020; Wang et al., 2021; He et al., 2020; Shi and Brasseur, 2020), the air quality improvement is much smaller than the expected (Shi et al., 2021; Diamond and Wood, 2020; Yan et al., 2022). Moreover, severe haze (COVID-19 haze) still occurred in northern China (Sulaymon et al., 2021; Le et al., 2020) and 

$\text{O}_3$ concentrations even showed significant increases (Zhang et al., 2021; Li et al., 2020). A number of studies were conducted to explain this air quality change by analyzing the effects of emission changes, meteorological variations and secondary production (Huang et al., 2021; Le et al., 2020; Hammer et al., 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 unknown emission changes during COVID-19 restrictions, the emission reduction scenarios that used to represent the COVID-19 shutdown effects varied among different studies and did not consider the spatial and temporal heterogeneity of the emission changes, leading to biases in the model simulation (Zhao et al., 2021; Li et al., 2021; Hammer et al., 2021; Zheng et al., 2021) and uncertainty in the quantification of the contributions of different factors.

Pioneer studies by Zheng et al. (2021) and Forster et al. (2020) have derived multi-air pollutant emissions from social activity data using a bottom-up method, but due to the lack of detailed social activity data, large uncertainties existed in their estimates. The meteorologically and seasonally driven variability of the concentrations of air pollutants also prohibit drawing fully quantitative conclusions on the changes of emissions based on observations alone (Levelt et al., 2022). The emission inversion technique, which takes advantage of the chemical transport model (CTM) and real-time observations, provides an attractive way to estimate the sector-specific and space-based emission changes during COVID-19 restrictions (Levelt et al., 2022), as shown in Zhang et al. (2020), Zhang et al. (2021), Feng et al. (2020) and Hu et al. (2022). However, these studies only inversed the emissions of single species (e.g., $\text{NO}_x$ and $\text{SO}_2$) without insights into multiple species. In view of this discrepancy, in this study we developed a multi-air pollutant inversion system to simultaneously estimated the multi-air pollutant emissions in China, including $\text{NO}_x$, $\text{SO}_2$, $\text{CO}$, $\text{PM}_{2.5}$ and $\text{PM}_{10}$ during the COVID-19 restrictions using an ensemble Kalman filter (EnKF) and surface observations from the China National Environmental Monitoring Centre (CNEMC). Subsequently, the inversed emission inventory was used to quantify the contributions of emission changes versus meteorology variations to the changes in $\text{PM}_{2.5}$ and $\text{O}_3$ concentrations over the NCP region during the COVID-19 restrictions.

2 Method and data

We developed a high-resolution multi-air pollutant inversion system to estimate the daily emissions of $\text{NO}_x$, $\text{SO}_2$, $\text{CO}$, $\text{PM}_{2.5}$ and $\text{PM}_{10}$ in China during the COVID-19 restrictions. This system uses the NAQPMS (Nested Air Quality Prediction
Modelling System) model as the forecast model and the EnKF coupled with the state argumentation method as the inversion method. It has the capabilities of simultaneous inversion of multi-air pollutant emissions at high temporal (daily) and spatial (15km) resolutions. An iteration inversion scheme was also developed in this study to address the large biases in the a priori emissions. In the following sections, we briefly introduce each component of the inversion system.

2.1 Chemical transport model and its configuration

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

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

2.2 Surface Observations

The hourly concentrations of NO2, SO2, CO, PM2.5 and PM10 from CNEMC were used in this study to estimate the emissions during COVID-19. The spatial distributions of these observation sites are shown in Fig. 1, which contains 1436 observation sites covering most regions of China. Before assimilation, outliers of observations were first filtered out using the automatic outlier detection method developed by Wu et al. (2018) to prevent the adverse effects of the outliers on data assimilation. Then, the hourly concentrations were averaged to the daily values for the inversions of daily emissions.

The observation error is one of the key inputs to the data assimilation, which together with the background error determine the relative weights of the observation and background values on the analysis. The observation error includes
measurement error and representativeness error. The measurement error of each species was designated according to the officially released documents of the Chinese Ministry of Ecology and Environmental Protection (HJ 193-2013 and HJ 654-2013, available at http://www.cnemc.cn/jcgf/dqfh/, last accessed on 22 Oct 2022), which is 5% for PM$_{2.5}$ and PM$_{10}$ and 2% for SO$_2$, NO$_2$ and CO. A representativeness error arises from the different spatial scales that the discrete observation data and 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

The EnKF coupled with the state augmentation method was used in this study to constrain the emissions of multiple species. EnKF is an advanced data assimilation method proposed by Evensen (1994) that features representation of the uncertainties of the model state by a stochastic ensemble of model realizations. It is easily implemented and can update the state variable by using the flow-dependent background error covariance matrix. In the state augmentation method, the emissions of different species, together with the concentrations of related species, were treated as the state variable and were updated according to the relationship between the emissions and concentrations of related species.

Since the source emission data over mainland China in HTAP_v2.2 inventory is obtained from the MIX inventory (Li et al., 2017b), uncertainties of emissions of different species, including PMF, PMC, BC, OC, NO$_x$, CO, SO$_2$, NH$_3$ and NMVOC (nonmethane volatile organic compounds), were obtained from Li et al. (2017b) and Streets et al. (2003), which were represented by an ensemble of perturbed emissions generated by multiplying the a priori emissions with a perturbation factor $\beta_{ls}$:

$$E_{ls} = \beta_{ls} \circ E_{a}^s, \quad i = 1, 2, \ldots, N_{ens}$$

where $E_{ls}$ represents the vector of the $ith$ member of perturbed emissions for species $s$, $E_{a}^s$ represents the a priori emissions for this species, $\circ$ denotes the schur product and $N_{ens}$ denotes the ensemble size. Thus, the adjustment of emissions is equivalent to the adjustment of perturbation factors.

Considering that emission uncertainty is the major contributor to the uncertainties in air quality modelling, especially during the COVID-19 period when emissions changed rapidly, uncertainties in chemical variables were obtained through ensemble simulations driven by perturbed emissions. The ensemble size was chosen as 50 to maintain the balance between the filter performance and computational cost. After the ensemble simulations, emissions of multiple species were updated using a deterministic form of EnKF (DEnKF) proposed by Sakov and Oke (2008), which is formulated by

$$\overline{x^b} = \overline{x^b} + P_e^bH^T(HP_e^bH^T + R)^{-1}(y^o - H\overline{x^b})$$  \hspace{1cm} (2)

$$\overline{x^b} = \frac{1}{N}\sum_{i=1}^{N}x^b_t; \quad x^b_t = x^b - \overline{x^b}$$  \hspace{1cm} (3)

$$P_e^b = \frac{1}{N-1}\sum_{i=1}^{N}X^b_t(X^b_t)^T$$  \hspace{1cm} (4)

where $x$ denotes the state variables; $b$ the background state (a priori); $a$ the analysis state (posteriori); $P_e^b$ the ensemble-estimated background error covariance matrix and $N$ the ensemble size. $y^o$ represents the vector of observations with an
error covariance matrix of \( \mathbf{R} \), \( \mathbf{H} \) is the linear observational operator that maps the \( m \)-dimensional state vector \( \mathbf{x} \) to a \( p \)-dimensional observational vector \( \mathbf{Hx} \). The state variables were defined as follows according to state augmentation method during the assimilation:

\[
\begin{align*}
\mathbf{x}_i &= [\mathbf{c}_i, \mathbf{\beta}_i]^T, i = 1, 2, \ldots, N_{\text{ens}} \\
\mathbf{c}_i &= [\text{PM}_{2.5}, \text{PM}_{10-2.5}, \text{NO}_2, \text{SO}_2, \text{CO}]_i \\
\mathbf{\beta}_i &= [\beta_{\text{PMF}}, \beta_{\text{BC}}, \beta_{\text{OC}}, \beta_{\text{PMC}}, \beta_{\text{NO}_2}, \beta_{\text{SO}_2}, \beta_{\text{CO}}]_i
\end{align*}
\]

where \( \mathbf{x}_i \) represents the \( i \)th member of the assimilated state variable, which consists of the fields of chemical variables \( \mathbf{c}_i \) and emission perturbation factors \( \mathbf{\beta}_i \). Detailed descriptions of the model state variables are summarized in Table 1. The use 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., 2018; Ma et al., 2019). Moreover, to prevent spurious correlations between non- or weakly related variables, similar to Ma et al. (2019) and Miyazaki et al. (2012), state variable localization was used during assimilation, with observations of one particular species only allowing us to update the emissions of the same species. The corresponding relationship between the chemical observations and adjusted emissions is summarized in Table 1. The PM\(_{2.5}\) observations were one exception and were used to update the emissions of PMF (fine mode unspeciated aerosol), BC (black carbon) and OC (organic carbon) since the observations of speciated PM\(_{2.5}\) were not available in this study. The lack of speciated PM\(_{2.5}\) observations may lead to uncertainties in the estimated emissions of PMF, BC and OC. Therefore, we only analyzed the emissions of PM\(_{2.5}\), which were the sum of the emissions of these three species. Similarly, only PM\(_{10}\) emissions were analyzed in this study, which includes the emissions of PM\(_{2.5}\) and PMC (coarse mode unspeciated aerosol).

Due to the strict control measures implemented during the last decades, the emissions in China decreased dramatically from 2010 to 2020, especially for SO\(_2\). Thus, there are large biases in the a priori estimates of emissions in China (Zheng et al., 2018), which would lead to incomplete adjustments of a priori emissions and degrade the performance of assimilation. Therefore, an iteration inversion scheme was developed in this study to address the large biases of SO\(_2\) emissions. As illustrated in Fig. 2, the main idea of the iteration inversion scheme is to update the ensemble mean of the state variable using the inversion results of the \( k \)th iteration and corresponding simulations. The state variable used in the \((k + 1)\)th iterations is written as follows:

\[
\mathbf{x}^{k+1} = [\mathbf{c}^k + \mathbf{c}^e, \mathbf{\beta}^k + \mathbf{\beta}^e, \mathbf{\beta}^e]^T
\]

where \( \mathbf{c}^k \) represents the simulation results using the inversed emissions of the \( k \)th iteration, \( \mathbf{c}^e \) represents the \( i \)th member of ensemble simulations with an ensemble mean of \( \mathbf{\bar{c}}^e \), \( \mathbf{\beta}^k \) represents the perturbation factors of the \( k \)th iteration, and \( \mathbf{\beta}^e \) represents the \( i \)th member of the ensemble of perturbation factors with a mean value of \( \mathbf{\bar{\beta}}^e \).

Using this method, the biases of a priori emissions were well addressed as exemplified in Fig. 3 for SO\(_2\) emissions. Due 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 errors (RMSE = 8.7–23.0 \( \mu \)g/m\(^2\)) in simulating SO\(_2\) concentration over all regions of China even after the assimilation (first iteration). The biases and errors continued to decrease with the increasing of iteration times till the fourth iteration in which...
there were no significant improvement in SO\textsubscript{2} simulations compared to those in third iteration. These results suggested that the iteration inversion method used in this study can well constrain the a priori emission with large biases and, in this application, conducting three iteration is enough for constraining the emission. Besides SO\textsubscript{2} emissions, the iteration inversion scheme was also applied to the emissions of other species.

To reduce the influences of random model errors (e.g., errors in meteorological inputs) on the estimation of the variation in emissions, a 15-day running average was performed on our daily inversion results after the inversion estimation. It should be noted that the COVID-19 restrictions were initiated during the Spring Festival of China, which would also influence the air pollutant emissions in China. However, the inversion method used in this study did not differentiate the contributions of the Spring Festival from the COVID-19 restrictions. Similarly, the effects of natural emission changes were not differentiated in this study, which would lead to uncertainty in quantifying the effects of the COVID-19 restrictions on air pollutant emissions.

2.4 Quantification of the effects of emission changes and meteorological variations

In previous studies, the meteorological-induced (MI) changes were usually determined by the CTM with a fixed emission input setting and a varying meteorological input. Then, the difference between the MI changes and total changes in air pollutant concentrations is defined as emission-induced (EI) changes. Another approach to estimate EI changes is to perform simulations with a fixed meteorological input setting and varying emission inputs. Then, the MI changes are defined as the difference between EI changes and total changes in air pollutant concentrations. Due to the nonlinear effects of atmospheric chemical systems, these two methods yield different results. Thus, both methods were used in this study to account for the nonlinear effects. The averaged results of these two methods are used to represent the impacts of emission changes and meteorological variation on the air quality changes during the COVID-19 restrictions. In total, three scenario experiments were designed based on our inversion results. The first scenario simulation used the varying meteorological and emission inputs from the P1 to P2 period, which represents the real-world scenario and is used to estimate the total changes in air pollutant concentrations induced by emissions and meteorological changes from the P1 to P2 period (BASE scenario). The second scenario experiment used the varying meteorological inputs but replaced the emissions during the P2 period with those during the P1 period, which was used to estimate the MI changes using the first method (MET change scenario). The third scenario experiment used the varying emissions input and replaced the meteorological input during the P2 period with that during the P1 period, which was used to estimate the EI changes using the second method (EMIS change scenario).

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

\[ M_{\text{MET change scenario}}^{\text{MI}} = \text{conc}_{p2,\text{MET change scenario}} - \text{conc}_{p1,\text{MET change scenario}} \]  

\[ E_{\text{MET change scenario}}^{\text{EI}} = \text{conc}_{p2,\text{BASE scenario}} - \text{conc}_{p1,\text{BASE scenario}} - M_{\text{MET change scenario}}^{\text{MI}} \]

where \( M_{\text{MET change scenario}}^{\text{MI}} \) represents the MI changes estimated based on the results from the MET change scenario, \( \text{conc}_{p1,\text{MET change scenario}} \) and \( \text{conc}_{p2,\text{MET change scenario}} \) represent the averaged concentrations of air pollutants during the
P1 and P2 periods under the MET change scenario, $EI_{\text{MET change scenario}}$ represents the EI changes estimated based on the results from the MET change scenario, and $\text{conc}_{p1,\text{BASE scenario}}, \text{conc}_{p2,\text{BASE scenario}}$ respectively represent the averaged concentrations of air pollutants during the P1 and P2 periods under the BASE scenario. Similarly, the MI and EI changes estimated based on the second method are formulated as follows:

$$EI_{\text{EMISSION change scenario}} = \text{conc}_{p2,\text{EMISSION change scenario}} - \text{conc}_{p1,\text{EMISSION change scenario}}$$

$$MI_{\text{EMISSION change scenario}} = \text{conc}_{p2,\text{BASE scenario}} - \text{conc}_{p1,\text{BASE scenario}} - EI_{\text{EMISSION change scenario}}$$

Then, the estimations from these two methods are averaged to estimate the contributions of meteorological change and emission change to the changes in PM$_{2.5}$ and O$_3$ concentrations during the COVID-19 lockdown:

$$MI = (MI_{\text{EMISSION change scenario}} + MI_{\text{MET change scenario}})/2$$

$$EI = (EI_{\text{EMISSION change scenario}} + EI_{\text{MET change scenario}})/2$$

$$\text{contri}_{\text{met}} = \frac{MI}{MI+EI} \times 100$$

$$\text{contri}_{\text{emis}} = \frac{EI}{MI+EI} \times 100$$

where $\text{contri}_{\text{met}}$ and $\text{contri}_{\text{emis}}$ represent the relative contributions (%) of the meteorological variations and emission changes to the changes in air pollutant concentrations.

### 3 Results

We estimated the multi-air pollutant emissions from 1 Jan to 29 Feb 2020 when the pandemic was at its most serious, and the effects of the COVID-19 restrictions were most profound in China. According to different control phases of COVID-19 and the timing of the Chinese Lunar New Year, the whole time period was divided into three periods: before lockdown (P1, January 1-20), lockdown (P2, January 21-February 9) and after back-to-work day (P3, February 10-29) to better characterize the emission changes during the COVID-19 restrictions. Additionally, we analyzed the emission changes in different regions of China, including the North China Plain (NCP), Northeast China (NE), Southeast China (SE), Southwest China (SW), Northwest China (NW) and Central regions (defined in Fig. 1) to investigate the responses of emissions to the COVID-19 restrictions in different regions.

#### 3.1 Validation of the inversion results

We firstly validate our inversion system by using a cross-validation, in which 20% of observation sites were withheld from the emission inversion and used as the validation datasets. Figure S1–6 showed the concentrations of different air pollutants in China from 1st Jan to 29th Feb 2020 obtained from observations at validation sites and simulations using a priori and a posteriori emission. Commonly used statistical evaluation indices, including correlation coefficient (R), mean bias error (MBE), normalized mean bias (NMB) and root of mean square error (RMSE) are summarized in Table S1. The validation results suggest that the posteriori simulation agreed well with the observed concentrations for all species.
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 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 -10–7.6% for CO (Table S4). RMSE values were also significantly reduced in the a posteriori simulation which were 9.1–32.2µg/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 CO. Moreover, the inversion emission considerably improved the fit to the observed time evolution of air pollutants’ concentrations. The R values were improved for all species in the a posteriori simulation that were up to 0.74–0.94 for PM$_{2.5}$, 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 results have excellent performance in representing the magnitude and variation of these species’ emission in China during COVID-19 restrictions. Model performance in simulating O$_3$ concentration is relatively poor compared to other species although improvement was remarkable in NCP, NE and SE regions. This would be due to that the emission of volatile 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

The control of COVID-19 began on 23rd January when the Chinese government declared the first level of national responses to public health emergencies, one day before the 2020 Chinese New Year Eve. Figure 4 shows the time evolution of the normalized emission anomaly for different species in China from 1st January to 29th February. The temporal variation in the emission varied largely between NO$_x$ and other species. Due to the combined effects of the Spring Festival and COVID-19 lockdown, NO$_x$ emissions decreased continuously at the beginning of January until approximately one week after the implementation of the COVID-19 lockdown, with estimated decreases in NO$_x$ emissions of up to 46.7% from the P1 to P2 period (Table 2). Subsequently, the NO$_x$ emissions stabilized with small fluctuations until the official back-to-work day when the NO$_x$ emissions began to increase due to easing of the control measures and the resumption of business. According to inversion estimation, NO$_x$ emissions recovered by 6.5% during the P3 period. These results indicate that the temporal variation in our estimated NO$_x$ emissions agreed well with the timing of the Spring Festival and different control stages of COVID-19. However, for other species (i.e., PM$_{2.5}$, PM$_{10}$, SO$_2$ and CO), although their emissions generally decreased from 1st January to the end of the 2020 Spring Festival holiday, they showed much smaller reductions than the NO$_x$ emissions. The emission reduction for these species was only approximately 7.9–12.1% (Table 2). This is consistent with the inversion results by Hu et al. (2022) who found that SO$_2$ emissions in China decreased only by 9.2% during COVID-19 lockdown. In addition, the emissions of these species quickly rebounded to their normal level just one week after the end of the Spring Festival holiday. As estimated by our inversion results, the SO$_2$ emissions recovered by 7.2% during the P3 period, which 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% higher, respectively, than those during the P1 period.
Similar results were found in different regions of China (Fig. 5 and Table 3), where the NO\textsubscript{x} emissions decreased much more than other species. In addition, unlike the uniform decreases in NO\textsubscript{x} emissions in different regions of China (~45%), there was apparent spatial heterogeneity in the emission changes in PM\textsubscript{2.5}, PM\textsubscript{10}, SO\textsubscript{2} and CO (Table 3 and Fig. 6). For example, from the P1 to P2 period, the PM\textsubscript{2.5} emissions decreased by over 20% in the Central region but only by 8.8% in the NE region. The PM\textsubscript{2.5} emissions even increased by 5.5% in the NCP region, possible due to the increased emissions from heating and fireworks (Dai et al., 2020). The SW and central regions exhibited relatively larger emission reductions for these 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 more prominent in the SE, central and SW regions (Fig. 5 and Fig. 7), where emissions recovered by 6.0–16.4% for NO\textsubscript{x}, 7.5–19.8% for SO\textsubscript{2}, 7.4–13.1% for CO, 12.3–47.7% for PM\textsubscript{2.5} and 28.6–135.9% for PM\textsubscript{10} (Table 3). This result is consistent with the earlier degradation of the response level to the COVID-19 virus (from the first level to the second or third level) over these regions (Table S2). In contrast, there were decreases in emissions in the NCP, NE and NW regions. PM\textsubscript{2.5} 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). Moreover, we found that the PM\textsubscript{10} emissions surged in the NW and central regions, where the PM\textsubscript{10} emissions during the P3 period were almost two times larger than those during the P2 period (Table 3). However, this finding may be related to the enhanced sandstorms over these two regions rather than the effects of returning to work, which demonstrates the necessity to consider changes in natural emissions during COVID-19 restrictions. Thus, to reduce the effects of natural emissions on our findings, the same analysis was performed for the emissions over east China (Fig. S7) where emissions were dominated by 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

Two explanations may help clarify the unbalanced emission changes between NO\textsubscript{x} and other species. First, the COVID-19 lockdown policy has led to dramatic decreases in transportation activities throughout China; however, as shown in Fig. 4, the relative contributions of the transportation sector to the emissions of SO\textsubscript{2} (2.4%), CO (18.5%), PM\textsubscript{2.5} (6.1%) and PM\textsubscript{10} (4.7%) are much smaller than those for NO\textsubscript{x} emissions (34.3%) (Zheng et al., 2018; Li et al., 2017a). Thus, the reduction in traffic activities can only substantially decrease NO\textsubscript{x} emissions. Reductions in CO emissions were relatively larger than those for SO\textsubscript{2} and PM\textsubscript{2.5} emissions, which is consistent with the relatively larger contributions of the transportation sector to CO emissions. PM\textsubscript{10} emissions showed the largest reductions among these four species, which is related in part to the reduced dust emissions due to shutting down of construction sites during the lockdown period (Li et al., 2020). Second, as shown in Fig. 4, the industrial and residential sectors are the major contributors to the anthropogenic emissions of SO\textsubscript{2}, CO, PM\textsubscript{2.5} and PM\textsubscript{10} in China, together contributing 77.6%, 78.3%, 86.5% and 86.3%, respectively, to their total emissions. The much smaller reductions of these species’ emissions were thus in line with the fact that there were no intentional restrictions on heavy industry during the COVID-19 restrictions. A large number of non-interruptible processes, such as steel, glass, coke, refractory, petrochemical, electric power, and especially heating, cannot be stopped during the COVID-19 lockdown. According to statistical data from the National Bureau of Statistics of China (Fig. S9), the industrial and power sectors did
not show similar reductions in their activity levels as those seen in the transportation sector. Power generation and steel production even showed increases in many provinces, which corresponds well with the emission increases over these regions. In addition, since people were required to stay at home, residential emissions were likely increased due to the increased energy consumption for heating or cooking. Therefore, our inversion results supported the views that the emissions of species related to industrial and residential activities did not decline much during the lockdown period, and that the COVID-19 lockdown policy was largely a traffic control measure with small influences on other sectors.

3.3 Investigation of air quality change over the NCP region during COVID-19 restrictions

Using the inversion results, we reassessed the environmental impacts of the COVID-19 restrictions on the air pollution over NCP region. The NCP region was chosen because it is the key target region of air pollution control in China and where unexpected COVID-19 haze occurred. A major caveat in previous studies that explored the impacts of COVID-19 lockdowns on air quality is the uncertainty in the emission changes during COVID-19 restrictions. The inversion results enable us to give a more reliable assessment of the environmental impacts of COVID-19 restrictions. Figure 8 shows the observed changes in PM$_{2.5}$ and O$_3$ concentrations over the NCP region from the P1 to P2 period. The observations showed consistent reductions in PM$_{2.5}$ concentrations over the NCP region (by 13.6 μg/m$^3$). However, substantial increases in PM$_{2.5}$ concentrations were observed in the Beijing area (by 31.2 μg/m$^3$). In contrast to the widespread reductions in PM$_{2.5}$ concentrations, the O$_3$ concentrations significantly increased over the whole NCP region (by 28.3 μg/m$^3$) and the Beijing area (by 16.8 μg/m$^3$). The simulations based on our inversion results reproduced the observed changes in PM$_{2.5}$ and O$_3$ concentrations over the NCP region well, although the increases in O$_3$ concentrations were relatively overestimated in the simulation (Fig. 7-8).

As detailed in Sect 2.4, the simulated changes in air pollutant concentrations before and after lockdown were decomposed into meteorological-induced (MI) changes and emission-induced (EI) changes through two different scenarios to account for the nonlinearity of the atmospheric chemical system. The meteorological variation dominated the changes in PM$_{2.5}$ concentrations over the NCP region (Fig. 9), which contributed 90% of the PM$_{2.5}$ reductions over most parts of the NCP region. Moreover, this variation made significant contributions (57.9%) to the increases in PM$_{2.5}$ concentrations over the Beijing area. This finding suggested that meteorological variations played an irreplaceable role in the occurrence of COVID-19 haze around the Beijing area. Compared with the meteorological conditions before lockdown (Fig. 10), there were increases in relative humidity over northern China, which facilitated the reactions for aerosol formation and growth. Wind speed also decreased over the Beijing area accompanied by an anomalous south wind, which facilitated aerosol accumulation and the transportation of air pollutants from the polluted industrial regions of the Hebei Province to Beijing. The increases in boundary layer height from the P1 to P2 period were also much smaller in the Beijing area than in other areas of the NCP. Thus, the Beijing area has exhibited distinct meteorological variations from other areas of the NCP region, which correspond well to the different changes in PM$_{2.5}$ concentrations over the Beijing area.
The emission changes contributed slightly to the PM$_{2.5}$ reductions over the NCP region (8.6%). This is because, on the one hand, the large reductions in NO$_x$ emissions (by 44.4%) only reduced nitrate by approximately 10–30% due to the nonlinear effects of chemical reactions (Fig. 11), and on the other hand, the emissions of primary PM$_{2.5}$ and its precursors from other sectors changed little during the COVID-19 restrictions (Table 3). The emission changes contributed more to the increased PM$_{2.5}$ concentrations over the Beijing area (42.1%). This is mainly associated with the increases in primary PM$_{2.5}$ emissions around the Beijing area, as seen in Fig. 6, possibly due to the increased emissions from firework during the Spring Festival over the rural area of Beijing (Dai et al., 2020). Therefore, our results suggested that the unexpected COVID-19 haze was mainly driven by unfavorable meteorological conditions together with small changes or even increases in primary 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. (2021), who suggested that enhanced secondary aerosol formation was the main driver of severe haze during the COVID-19 restrictions. To investigate it, we further analyzed the changes in the concentrations of secondary inorganic aerosols (SIAs). First, we evaluated our model results against the observed SIA concentrations, which showed that the model results using our inversion emissions well reproduced the observed concentrations of SIAs over the NCP region (Fig. 12) with mean bias (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 SIA concentrations over the Beijing area, especially for sulfate concentrations, were also captured in our simulations (Fig. 11), although underestimation occurred due to the uncertainty in simulating SIA concentrations. Through sensitivity experiments, we found that the increases in SIA concentrations were still driven by meteorological variations (Fig. 13). In fact, the emission reductions only led to a 10% decrease in SIA concentrations over the NCP region. This finding suggests that the enhanced secondary aerosol formation was likely mainly driven by the unfavorable meteorological conditions associated with higher temperature and relative humidity instead of the emission reductions during the lockdown period. This is in line with the observation evidences from Ma, T et al (2022) who emphasized that the increased temperature and relative humidity promoted the formation of secondary pollutants during the COVID-19 restrictions.

In terms of O$_3$ concentrations, the emission changes subsequently became the dominant contributor to the O$_3$ increases by more than 100% in the Beijing area and by 96.0% over the NCP region. This result is mainly because the lockdown period occurred in midwinter when photochemical O$_3$ formation was minimal; thus, the large increase in O$_3$ is expected solely from the effect of the reduced titration reaction associated with the large reductions in NO$_x$ emissions. Although the higher temperature and slower wind speed during the lockdown period were favorable for the increases in O$_3$ concentrations, their contributions were much smaller than those of emission changes (Fig. 9). These results suggested that control measures, such as COVID-19 restrictions, were inefficient for air pollution mitigation in China considering the high economic cost of the COVID-19 restrictions.
4 Conclusions

The COVID-19 pandemic is an unprecedented event that significantly influenced the social activity and associated emissions of air pollutants. Our results provide a quantitative assessment of the influences of COVID-19 restrictions on multi-air pollutant emissions in China. Otherwise, understanding of the relationship between air quality and human activities may be biased. The inversion results provide important evidences that the COVID-19 lockdown policy was largely a traffic control measure with substantially reducing impacts on NO\textsubscript{x} emissions but much smaller influences on the emissions of other species and other sectors. Traffic control has widely been considered to be the normal protocol in implementing regulations in many cities of China, but its effectiveness on air pollution control is still disputed (Han and Naeher, 2006; Zhang et al., 2007; Chen et al., 2021; Cai and Xie, 2011; Chowdhury et al., 2017; Li et al., 2017c). Thus, the COVID-19 restrictions provided us with a real nationwide traffic control scenario to investigate the effectiveness of traffic control on the mitigation of air pollution in China. The results suggested that traffic control as a separate pollution control measure has limited effects on the coordinated control of high concentrations of O\textsubscript{3} and PM\textsubscript{2.5} under the current air pollution conditions in China. In this case, the PM\textsubscript{2.5} concentrations were slightly reduced, while leading to substantial increases in O\textsubscript{3} concentrations. Severe haze was also not avoided during the COVID-19 restrictions due to unbalanced emission changes from other sectors and unfavorable meteorological conditions. China is now facing major challenges in both controlling PM\textsubscript{2.5} and controlling emerging O\textsubscript{3} pollution. The tragic COVID-19 pandemic has revealed the limitation of the road traffic control measure in the coordinated control of PM\textsubscript{2.5} and O\textsubscript{3}. More comprehensive regulations for multiple precursors from different sectors are required in the future to address O\textsubscript{3} and PM\textsubscript{2.5} pollution in China.
Table 1. Corresponding relationship between the chemical observations and adjusted emissions

<table>
<thead>
<tr>
<th>Species</th>
<th>Descriptions</th>
<th>Observations that used for inversions of this species</th>
</tr>
</thead>
<tbody>
<tr>
<td>BC</td>
<td>black carbon</td>
<td>PM$_{2.5}$</td>
</tr>
<tr>
<td>OC</td>
<td>organic carbon</td>
<td>PM$_{2.5}$</td>
</tr>
<tr>
<td>PMF</td>
<td>fine mode unspeciated aerosol</td>
<td>PM$_{2.5}$</td>
</tr>
<tr>
<td>PMC</td>
<td>coarse mode unspeciated aerosol</td>
<td>PM$<em>{10} - PM</em>{2.5}$</td>
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<tr>
<td>NO$_x$</td>
<td>nitrogen oxide</td>
<td>NO$_2$</td>
</tr>
<tr>
<td>SO$_2$</td>
<td>sulfur dioxide</td>
<td>SO$_2$</td>
</tr>
<tr>
<td>CO</td>
<td>carbon monoxide</td>
<td>CO</td>
</tr>
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</table>

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

<table>
<thead>
<tr>
<th></th>
<th>NO$_x$</th>
<th>SO$_2$</th>
<th>CO</th>
<th>PM$_{2.5}$</th>
<th>PM$_{10}$</th>
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<tr>
<td>P1 (Gg/day)</td>
<td>72.9</td>
<td>23.8</td>
<td>1160.2</td>
<td>44.5</td>
<td>75.5</td>
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<tr>
<td>P2 (Gg/day)</td>
<td>41.9</td>
<td>21.5</td>
<td>1037.4</td>
<td>40.9</td>
<td>66.4</td>
</tr>
<tr>
<td>P3 (Gg/day)</td>
<td>44.8</td>
<td>23.2</td>
<td>1078.2</td>
<td>45.9</td>
<td>108.4</td>
</tr>
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<td>(P2-P1)/P1</td>
<td>-42.5%</td>
<td>-9.7%</td>
<td>-10.6%</td>
<td>-7.9%</td>
<td>-12.1%</td>
</tr>
<tr>
<td>(P3-P2)/P1</td>
<td>3.9%</td>
<td>7.2%</td>
<td>3.6%</td>
<td>11.2%</td>
<td>55.7%</td>
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<tr>
<td>(P3-P1)/P1</td>
<td>-38.6%</td>
<td>-2.5%</td>
<td>-7.0%</td>
<td>3.3%</td>
<td>43.6%</td>
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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

<table>
<thead>
<tr>
<th>Region</th>
<th>NO$_x$</th>
<th>PM$_{2.5}$</th>
<th>PM$_{10}$</th>
<th>SO$_2$</th>
<th>CO</th>
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<td>NCP</td>
<td>(P2-P1)/P1</td>
<td>-44.4%</td>
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<td></td>
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<td>(P3-P1)/P1</td>
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<td>-8.8%</td>
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<td>-3.2%</td>
</tr>
<tr>
<td></td>
<td>(P3-P2)/P1</td>
<td>-6.0%</td>
<td>-19.2%</td>
<td>23.7%</td>
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</tr>
<tr>
<td></td>
<td>(P3-P1)/P1</td>
<td>-47.8%</td>
<td>-28.0%</td>
<td>20.2%</td>
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</tr>
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<td>SE</td>
<td>(P2-P1)/P1</td>
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<td>-9.5%</td>
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<td></td>
<td>(P3-P2)/P1</td>
<td>10.2%</td>
<td>12.3%</td>
<td>28.6%</td>
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<td>-12.6%</td>
<td>-25.9%</td>
<td>-17.5%</td>
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<tr>
<td></td>
<td>(P3-P1)/P1</td>
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<tr>
<td>NW</td>
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<td>(P3-P2)/P1</td>
<td>-21.1%</td>
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<td></td>
<td>(P3-P1)/P1</td>
<td>-59.6%</td>
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<td>Central</td>
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<td>18.5%</td>
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<td>(P3-P1)/P1</td>
<td>-27.4%</td>
<td>0.7%</td>
<td>120.3%</td>
<td>7.9%</td>
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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.
Figure 2: Illustration of the iteration inversion scheme used in this study.
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.
Figure 4: (a) Time series of normalized emission anomalies estimated by inversion results for different species in China from 1st January to 29th February 2020, and (b-f) Relative contributions of different sectors to the total anthropogenic emissions of NO\textsubscript{x}, PM\textsubscript{2.5}, PM\textsubscript{10}, CO and SO\textsubscript{2} obtained from Zheng et al. (2018). The normalized emission anomaly is calculated by the emission 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\textsubscript{x}, (b) SO\textsubscript{2}, (c) CO, (d) PM\textsubscript{2.5} and (e) PM\textsubscript{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) PM$_{2.5}$ and (b, d) O$_3$ over the NCP region from the pre-lockdown period (P1) to the lockdown period (P2).
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.
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.
Figure 12: Time series of observed and simulated concentrations of \( \text{NO}_3 \), \( \text{SO}_4 \) and \( \text{NH}_4 \) in (a-c) Beijing, (b-f) Tianjin, (g-i) Hebei, (j-l) Henan and (m-o) Shangdong province from 1st January to 29th February 2020.
Figure 13: Meteorology-induced (MI) changes in the concentrations of (a) ANO$_3$, (b) ASO$_4$ and (c) ANH$_4$, as well as Emission-induced (EI) changes in the concentrations of (d) ANO$_3$, (e) ASO$_4$ and (f) ANH$_4$.

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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