



Airborne hazardous trace metals pose significant risks to human health. However, the response characteristics of ambient trace metals to emission reductions remain poorly understood. The COVID-19 pandemic offered a unique opportunity to investigate these response mechanisms and optimize emission control strategies. In this study, we employed the GEOS-Chem chemical transport model to predict global variations in atmospheric concentrations of nine hazardous trace metals (As, Cd, Cr, Cu, Mn, Ni, Pb, V, and Zn) and assess their responses to COVID-19 lockdown measures. Our results revealed that global average concentrations of As, Cd, Cr, Cu, Mn, Ni, and V decreased by 1-7%, whereas Pb and Zn levels increased by 1% and 2%, respectively. The rise in Pb and Zn concentrations during lockdowns was primarily linked to sustained coal combustion and non-ferrous smelting activities, which remained essential for residential energy demands. Spatially, India, Europe, and North America experienced the most pronounced declines in trace metal levels, while Sub-Saharan Africa and Australia showed minimal sensitivity to lockdown-induced emission



30 reductions. Based on the scenario analysis, we found the concentrations of trace metals displayed  
31 linear response to emission reduction. Combined with the health risk assessment, we demonstrated  
32 the reduced emissions of Pb and As during the lockdown period yielded the greatest health  
33 benefits—Pb reductions were associated with lower non-carcinogenic risks, while As declines  
34 contributed most significantly to reduced carcinogenic risks. Targeting fossil fuel combustion  
35 should be prioritized in Pb and As mitigation strategies.

## 36 1. Introduction

37 With the rapid advancement of industrial development and urbanization, numerous hazardous  
38 trace metals have been released into the atmosphere (Cheng et al., 2015; Yu et al., 2012; Zhu et al.,  
39 2020). These toxic elements injected into the air can negatively impact terrestrial and aquatic  
40 ecosystems through dry or wet deposition and pose significant risks to human health through long-  
41 term exposure (Al-Sulaiti et al., 2022; Pan and Wang, 2015; Sharma et al., 2023). Certain trace  
42 elements, including arsenic (As), cadmium (Cd), chromium (Cr), and lead (Pb), have been classified  
43 as carcinogens by the International Agency for Research on Cancer (IARC) (Bai et al., 2023; Loomis  
44 et al., 2018; Pearce et al., 2015). Therefore, it is crucial to investigate the spatiotemporal  
45 characteristics of hazardous trace metals in the atmosphere and assess their health impacts. Such  
46 efforts are instrumental in identifying hotspots and formulating effective control measures to  
47 mitigate health risks.

48 A growing body of studies have explored the long-term trends of ambient trace elements in  
49 various cities (Das et al., 2023; Guo et al., 2022; Nirmalkar et al., 2021). For example, Farahani et  
50 al. (2021) reported significant decreases in PM<sub>2.5</sub>-bound levels of nickel (Ni), vanadium (V), zinc  
51 (Zn), lead (Pb), manganese (Mn), and copper (Cu) in central Los Angeles between 2005 and 2018  
52 (Farahani et al., 2021). Very recently, Li et al. (2022) confirmed that concentrations of Pb, zinc (Zn),  
53 and arsenic (As) in PM<sub>2.5</sub> in Tangshan decreased by 62%, 59%, and 54% from 2017 to 2020,  
54 respectively, owing to the implementation of clean air actions. Although some studies have analyzed  
55 the long-term trends of ambient hazardous trace metals in specific regions, the spatiotemporal  
56 variations in most areas remain poorly understood due to the limited spatial representativeness of  
57 monitoring sites. To address these limitations, some researchers have utilized chemical transport  
58 models to predict regional or global concentrations of ambient hazardous trace metals. For instance,



59 Liu et al. (2021) employed the CMAQ model to estimate the concentrations of 11 trace elements  
60 and found that most of these elements exhibited higher levels in the North China Plain (NCP).  
61 Additionally, Zhang et al. (2020) used the GEOS-Chem model to predict global ambient arsenic  
62 (As) concentrations from 2005 to 2015. Their findings revealed that atmospheric As levels in India  
63 exceeded those in eastern China, primarily due to the sharp increase in coal combustion across India.  
64 However, to date, the spatiotemporal variations of multiple hazardous trace metals on a global scale  
65 remain poorly understood. Moreover, the response of ambient trace elements to changes in  
66 emissions is still unknown. Bridging this knowledge gap is crucial for implementing effective  
67 control and prevention measures targeted at specific trace elements.

68 COVID-19 swept across the globe between January and April 2020 (Fonseca et al., 2021;  
69 Saadat et al., 2020; Venter et al., 2020). To prevent the rapid spread of this complex disease, many  
70 strict lockdown measures were implemented, including the partial or complete closure of  
71 international borders, the shutdown of nonessential businesses, and restrictions on citizen mobility  
72 (Meo et al., 2020; Onyeaka et al., 2021; Xing et al., 2021). These stringent control measures led to  
73 dramatic decreases in the levels of many gaseous precursors, such as  $\text{NO}_x$  and CO. Keller et al.  
74 (2021) estimated that global  $\text{NO}_x$  emissions were reduced by 3.1 Tg N during January-June 2020,  
75 accounting for 5.5% of total anthropogenic emissions. In addition, many studies have analyzed the  
76 response mechanisms of secondary pollutants to emission reductions during this period. Li et al.  
77 (2023b) identified three distinct response mechanisms of secondary nitrogen-bearing components  
78 to the reduction in  $\text{NO}_x$  emissions. Although the impacts of COVID-19 lockdown on multiple  
79 pollutants have been explored, the response of ambient hazardous trace metals to emission changes  
80 during the COVID-19 period still remains unknown. Additionally, the separate contributions of  
81 emission reductions and meteorological factors to ambient hazardous trace metal levels are also  
82 unclear. The COVID-19 pandemic provided an unprecedented opportunity to uncover these  
83 response mechanisms and marginal benefits of emission reduction, offering valuable insights for  
84 developing effective air pollution mitigation strategies.

85 Here, we developed a new trace metal emission inventory and employed the GEOS-Chem  
86 model to predict the global variations of ambient trace elements before and during the COVID-19  
87 period. First, we analyzed the spatiotemporal variations of ambient trace metal levels. Then, we



distinguished the contributions of emission changes and meteorology. Finally, we quantified the health benefits resulting from emission reductions. Our study provides valuable insights and targeted policy implications for future air pollution control.

## 2. Materials and methods

### 2.1 The global emission inventory of hazardous trace metals

According to the classification standard proposed by Streets et al. (2011), all countries and regions worldwide could be categorized into five groups, ranging from the most developed (Region 1) to the least developed (Region 5). Anthropogenic emission factors and the removal efficiency of hazardous trace metals vary significantly among these regions. Detailed data were obtained from Zhu et al. (2020). Coal combustion sources were further divided into two subcategories: coal-fired power plants and other coal-fired sectors. Non-coal combustion sources were classified into six subsectors: liquid fuel combustion, ferrous metal smelting, non-ferrous metal smelting, non-metallic mineral manufacturing, vehicle emissions, and municipal solid waste incineration. Natural emissions, including those from dust, biomass burning, and sea salt, were estimated in detail using the methodology described by Wu et al. (2020). Based on these categorizations and methodologies, an emission inventory for nine trace metals including As, Cd, Cr, Cu, Mn, Ni, Pb, V, and Zn was developed. The detailed emission inventory description was introduced in Supporting Information.

### 2.2 Ground-level observations

Most of the ground-level ambient hazardous trace metal observations ( $PM_{10}$ ) mainly focused on China, Western Europe, and Contiguous United States. China did not possess regular ground-level observations of ambient trace metals, and thus we only collected these data from previous references. Both of Western Europe and Contiguous United States possessed regular ambient trace metal observations. The European Monitoring and Evaluation Programme (EMEP) comprises of more than 100 sites about trace metal observations across Europe in past 20 years. The quality control of EMEP was explained by Tørseth et al., (2012). The dataset of daily ambient trace metal concentrations in many sites across the United States were downloaded from the website of <https://www.epa.gov/> (Figure S1).

### 2.3 GEOS-Chem model

The GEOS-Chem model (v12-01) was utilized to simulate the concentration differences of nine



hazardous trace metals in the atmosphere during the period from January 23 to April 30, comparing 2020 to 2019. These differences were attributed to the combined effects of emission changes and meteorological variations. The core mechanism of this model integrates tropospheric NO<sub>x</sub>-VOC-O<sub>3</sub>-aerosol chemistry (Mao et al., 2010; Park et al., 2004). Wet deposition processes include sub-grid scavenging in convective updrafts, in-cloud rainout, and below-cloud washout (Liu et al., 2001). Dry deposition was calculated using a resistance-in-series model (Wesely, 2007). The model was driven by meteorological data assimilated from the MERRA2 reanalysis (Qiu et al., 2020). A global simulation at a  $2 \times 2.5^\circ$  resolution was conducted to estimate the concentrations of hazardous trace metals on a global scale (Qiu et al., 2020). Anthropogenic trace metal emission inventories for 2019 and 2020 were derived from Section 2.1, while natural emissions included dust, biomass burning, and sea salt. The modelled trace metal concentration is the sum of TE concentrations in particle sizes  $\leq 2.5 \mu\text{m}$  in diameter (accumulation mode) and particle sizes between 2.5 and  $10 \mu\text{m}$  in diameter (coarse mode). Model performance was evaluated using several statistical indicators, with the detailed equations provided in the Supplementary Information (SI).

To isolate the effects of emission reductions and meteorological changes on ambient hazardous trace metals, simulations using the 2019 emission inventory and 2020 meteorological conditions ( $GC_{2019\text{emi}-2020\text{met}}$ ) were also conducted.  $GC_{2020}$  and  $GC_{2019}$  represent simulation results based on the emissions and meteorological conditions of 2020 and 2019, respectively. The pollutant concentration differences between 2019 and 2020 caused by emissions ( $GC_{\text{emi}}$ ) and meteorology ( $GC_{\text{met}}$ ) were quantified using the following equations:

$$GC_{\text{emi}} = GC_{2020} - GC_{2019\text{emi}-2020\text{met}} \quad (1)$$

$$GC_{\text{met}} = GC_{2019\text{emi}-2020\text{met}} - GC_{2019} \quad (2)$$

The detailed calculation process is as follows. First, the concentrations of ambient trace metals for both 2019 and 2020 were simulated. In the second step, the meteorology for 2020 was fixed, and the emission inventory was adjusted to 2019 to calculate  $GC_{2019\text{emi}-2020\text{met}}$ . The simulated ambient trace metal levels for 2020 were then subtracted from the simulated concentrations with 2019 emissions and 2020 meteorology (Eq. 1) to determine the contribution of emissions to the total difference in ambient trace metals during the COVID-19 period ( $GC_{\text{emi}}$ ). Similarly, based on Eq. 2, the same method was applied to estimate the contribution of meteorology to the total difference in



145 ambient trace metals during the COVID-19 period ( $GC_{met}$ ). Different regions exerted lockdown  
146 measures during different periods. The lockdown periods in various regions were collected from  
147 [https://en.wikipedia.org/wiki/COVID-19\\_pandemic\\_lockdowns](https://en.wikipedia.org/wiki/COVID-19_pandemic_lockdowns) (Keller et al., 2021). Five scenarios  
148 including base, 20%\_reduction, 40%\_reduction, 60%\_reduction, and 80%\_reduction were set up to  
149 assess the response of trace metal concentrations to emission change. For example, the  
150 20%\_reduction means the global trace metal emission experienced 20% reduction.

#### 151 2.4 Health risk assessment model

152 In this study, the carcinogenic and non-carcinogenic risks associated with hazardous trace  
153 metals in aerosols were evaluated using statistical thresholds established by IARC. Based on the  
154 IARC classification, trace metals such as As, Cd, Cr, Cu, Mn, Ni, Pb, V, and Zn are identified as  
155 potentially carcinogenic to humans (Marufi et al., 2024; Tikadar et al., 2024). The risks of exposure  
156 to these hazardous trace metals were assessed for both adults ( $\geq 24$  years old) and children ( $< 6$   
157 years old) using carcinogenic risk (CR) and hazard quotient (HQ) metrics. Both CR and HQ were  
158 derived from the average daily dose (ADD). The formulas used to calculate ADD, CR, and HQ were  
159 adopted from Kan et al. (2021) and Zhang et al. (2021) (Table S1-S2):

$$160 \quad \text{ADD} = (C \times \text{InhR} \times \text{EF} \times \text{ED}) / (\text{BW} \times \text{AT}) \quad (3)$$

$$161 \quad \text{HQ} = \text{ADD} / \text{RfD} \quad (4)$$

$$162 \quad \text{CR} = \text{ADD} \times \text{CSF} \quad (5)$$

163 where  $C$  ( $\text{mg m}^{-3}$ ) represents the concentration of hazardous trace metals in the atmosphere, whereas  
164  $\text{InhR}$  denotes the respiratory rate ( $\text{m}^3 \text{d}^{-1}$ ) (Table S1).  $\text{EF}$  refers to the annual exposure frequency ( $\text{d}$   
165  $\text{y}^{-1}$ ),  $\text{ED}$  is the exposure duration (years),  $\text{BW}$  represents the average body weight (kg), and  $\text{AT}$  is  
166 the average exposure time (days).  $\text{ADD}$  indicates the average daily intake ( $\text{mg kg}^{-1} \text{d}^{-1}$ ) of hazardous  
167 trace metals,  $\text{RfD}$  is the reference dose ( $\text{mg kg}^{-1} \text{d}^{-1}$ ) derived from reference concentrations, and  $\text{CSF}$   
168 is the cancer slope factor ( $\text{kg d mg}^{-1}$ ). The potential non-carcinogenic risk of hazardous trace metals  
169 is considered high if the hazard quotient (HQ) exceeds 1.0, indicating significant health concerns.  
170 Conversely, an HQ below 1.0 suggests negligible health risks. The carcinogenic risk (CR) of each  
171 hazardous trace metal is assessed as significant if CR exceeds  $10^{-4}$ .

### 172 3. Results and discussions

#### 173 3.1 Model evaluation



174 Initially, the GEOS-Chem model was employed to estimate ambient trace metal concentrations  
175 at the global scale. The correlation coefficients (R values) between the simulated and observed  
176 values for As, Cd, Cr, Cu, Mn, Ni, Pb, V, and Zn were 0.76, 0.75, 0.81, 0.75, 0.79, 0.81, 0.83, 0.78,  
177 and 0.79 (Figure 1), respectively. The root mean square errors (RMSE) for As, Cd, Cr, Cu, Mn, Ni,  
178 Pb, V, and Zn were 2.57, 4.45, 4.84, 11.1, 12.7, 3.64, 28.1, 9.33, and 30.1 ng/m<sup>3</sup>, respectively. The  
179 mean absolute errors (MAE) for these trace metals were 0.92, 2.89, 2.15, 5.53, 6.73, 1.60, 12.3, 4.09,  
180 and 13.5 ng/m<sup>3</sup>, respectively. Only Ni and V were overestimated, while the concentrations of most  
181 other trace metals were slightly underestimated. The predictive R values in our study were generally  
182 higher than those reported by Liu et al. (2021), with the exception of Mn (R = 0.87) and Cu (R =  
183 0.86). It is likely that Liu et al. (2021) simulated ambient trace metal concentrations specifically in  
184 China, which experiences some of the most severe trace metal pollution globally. As a result, the  
185 simulated values in China were often underestimated. In contrast, our study conducted global  
186 simulations, which helped avoid such significant underestimation. Additionally, the predictive  
187 accuracy for ambient As concentrations in our study was slightly higher than that of Zhang et al.  
188 (2020) (R = 0.69). Overall, the predictive accuracy of ambient trace metal concentrations in our  
189 study was satisfactory, allowing us to use these data to further analyze the spatiotemporal  
190 characteristics of trace metal distributions.

### 191 3.2 The concentration differences and health benefits of ambient trace metals during 2019 and 2020

192 Based on the simulated results, the global average concentrations of ambient As, Cd, Cr, Cu,  
193 Mn, Ni, Pb, V, and Zn during January-April in 2019 (2020) were 0.05 (0.04), 0.02 (0.02), 0.12 (0.12),  
194 0.39 (0.37), 0.21 (0.20), 0.17 (0.16), 0.82 (0.83), 0.24 (0.22), and 0.43 (0.44) ng/m<sup>3</sup> (Figure 2 and  
195 Figure S2-S4), respectively. Most trace metals in the atmosphere experienced decreases during the  
196 COVID-19 period in 2020 compared to the "business-as-usual" period in 2019. The global average  
197 concentrations of ambient As, Cd, Cr, Cu, Mn, Ni, and V decreased by 5%, 1%, 1%, 4%, 5%, 7%,  
198 and 7%, respectively. However, global average concentrations of particulate Pb and Zn exhibited  
199 slight increases of 1% and 2%, respectively, during the same period.

200 The ambient hazardous trace metals exhibited distinct spatial variations across regions during  
201 this period. Nearly all trace metal concentrations in India (Ind), Western Europe (WE), North  
202 America (NA), and Russia (Rus) showed significant decreases following the COVID-19 outbreak



(Figure 3). For example, particulate As concentrations in these regions decreased by 4%, 17%, 10%, and 12%, respectively. These reductions are likely linked to the high trace metal emissions during the "business-as-usual" period, driven by fossil fuel combustion for industrial activities (activity level decreased by 23%), which were substantially curtailed by stay-at-home orders (Bai et al., 2023; Doumbia et al., 2021). In China, the concentrations of As, Cu, Mn, Ni, and V decreased by 3%, 2%, 3%, 3%, and 4%, respectively. However, ambient levels of Cd, Cr, Pb, and Zn increased by 5%, 2%, 6%, and 8%, respectively. It was assumed that most of these trace metals were mainly derived from residential fossil fuel combustion (increase by 5%) and essential industrial emissions (remained relatively stable) (Zhu et al., 2020; Zhu et al., 2018), which even increased due to the stay-at-home order. In South America (SA), most trace metals, except for As and Mn, showed slight decreases during the COVID-19 period. Conversely, ambient trace metal concentrations in Sub-Saharan Africa (SS) and Australia (Aus) were relatively insensitive to COVID-19 lockdown measures, remaining stable throughout the period. It was supposed that the concentrations of trace metals in these regions were relatively low during the "business-as-usual" period.

Although the absolute concentrations provide an overall indication of the impact of lockdown measures on ambient trace metals, the contribution of meteorological factors cannot be disregarded. Meteorological conditions can significantly influence ambient trace metal concentrations, potentially complicating the interpretation of trace metal responses to emission changes. To isolate this effect, we applied the isolation technique to separate the contributions of emission changes from those of meteorological factors. At the global scale, most ambient trace metal concentrations, except for Pb and Zn, showed slight decreases, reflecting minor emission reductions coupled with favorable meteorological conditions that decreased concentrations (Figure 4 and Figure S5-S12). However, the slight increases in Pb and Zn concentrations were likely due to the fact that favorable meteorological conditions could not offset the significant rise in emissions of these metals during the period. It is assumed that Pb and Zn were primarily derived from coal combustion and non-ferrous smelting industries (Li et al., 2023a). In our study, the global residential energy consumption even increased by 5% during COVID-19 period based on the statistical data (<https://www.iea.org/>), which could lead to slight increases of Pb and Zn concentrations. Previous studies also have confirmed that residential coal consumption increased notably (10-20%) during the lockdown





232 period (Li et al., 2021; Smith et al., 2021). Emission changes and meteorological conditions  
233 exhibited significant spatial heterogeneity. In China, overall meteorological conditions favored trace  
234 metal removal, though unfavorable conditions in northern Hebei Province and Northeast China  
235 during January 2020 limited this effect (Chang et al., 2020; Huang et al., 2020; Li et al., 2023b).  
236 Most regions, such as South China and West China, experienced favorable meteorological  
237 conditions, as confirmed by several studies (Li et al., 2023b). Despite this, emission-induced  
238 concentrations of Cd, Cr, Pb, and Zn in China still showed slight increases during the COVID-19  
239 period, suggesting that essential sectors, such as coal and non-ferrous industries, could not be fully  
240 shut down. The data of National Bureau of Statistics (<https://data.stats.gov.cn/index.htm>) also  
241 verified that the coal consumption and products of non-ferrous industries in China nearly remained  
242 invariable during this period. In regions like India, WE, NA, and Russia, favorable meteorological  
243 conditions combined with substantial emission reductions led to decreases in most trace metal  
244 concentrations. Keller et al. (2021) demonstrated that stay-at-home orders in WE and parts of NA  
245 resulted in more than a 50% reduction in NO<sub>x</sub> emissions (mainly from vehicle emission and  
246 industrial activity), a trend consistent with the trace metal reductions observed in our study. In SA,  
247 the emission-induced reductions of trace metals were more pronounced than those due to  
248 meteorological conditions. This might be because countries such as Chile and Brazil have relatively  
249 high non-essential trace metal emissions, which were strongly affected by lockdown measures  
250 (Huber et al., 2016; La Colla et al., 2021; Zhu et al., 2020). In SS, emission-induced reductions also  
251 exceeded meteorological effects for most trace metals, except for Mn and Ni. However, in Australia,  
252 emission-induced reductions were less significant compared to SA and SS. Despite its developed  
253 mineral mining industries, Australia has more effective pollution control measures, resulting in  
254 lower total trace metal emissions (Zhu et al., 2020; Pacyna and Pacyna, 2001; Zhou et al., 2015).

255 In order to assess the response of trace metal concentrations to assumed emission change, the  
256 five emission scenarios (base (annual average concentration in 2019), 20%, 40%, 60%, and 80%)  
257 were set up to evaluate the impact of emission reduction. In our study, the results suggested that the  
258 As concentrations in China, India, WE, NA, SA, SS, Russia, and Australia decreased from 1.14,  
259 0.55, 0.13, 0.09, 0.04, 0.01, 0.08, and 0.01 to 0.23, 0.11, 0.03, 0.02, 0.01, 0, 0.02, and 0 ng/m<sup>3</sup>  
260 (Figure S13a), respectively. Besides, concentrations of other trace metals also displayed linear



261 decreases with the increasing of emission reduction ratio (Figure S13). It was assumed that these  
262 trace metals did not participate in chemical reactions in the atmosphere and were only affected by  
263 physical processes.

### 264 3.3 Unexpected health benefits from COVID-19 period

265 Based on the equations of (3)-(5), the global health risk indicators including CR (carcinogenic  
266 risk) and HQ (hazard quotient) values were calculated. At the global scale, among all of the trace  
267 metals, Pb showed the highest CR values ( $9.7 \times 10^{-8}$  and  $2.4 \times 10^{-7}$ ), followed by Ni ( $3.5 \times 10^{-8}$  and  
268  $8.6 \times 10^{-8}$ ), Cd ( $3.0 \times 10^{-8}$  and  $7.2 \times 10^{-8}$ ), As ( $1.6 \times 10^{-8}$  and  $3.9 \times 10^{-8}$ ), and Cr ( $1.4 \times 10^{-8}$  and  $3.4 \times 10^{-8}$ ),  
269 and the lowest ones for Cu, Mn, V, and Zn in both of 2019 and 2020. For non-carcinogenic risk, HQ  
270 values showed the higher values for As ( $3.5 \times 10^{-5}$  and  $9.0 \times 10^{-5}$ ) and Cu ( $2.2 \times 10^{-5}$  and  $5.5 \times 10^{-5}$ ).  
271 The results were in good agreement with previous studies because As often showed the higher non-  
272 carcinogenic risk (Li et al., 2023).

273 The CR and HQ values can be summed to estimate the total carcinogenic and non-carcinogenic  
274 risks. By comparing the CR and HQ values between 2019 and 2020, we quantified the health burden  
275 (or benefits) attributable to the COVID-19 lockdown (Figure 5 and Figure S14-S15). At the spatial  
276 scale, significant decreases in health burden were observed across most regions of the NCP,  
277 Southeast China, WE, the Dun River Basin, and the eastern United States. Following January 23,  
278 2020, at least 29 provinces in China reported confirmed COVID-19 cases. In response, strict control  
279 measures, including the shutdown of commercial activities and the lockdown of entire cities, were  
280 implemented. These measures significantly reduced emissions, particularly in populous regions  
281 such as the NCP and Southeast China (Jia et al., 2021; Shi and Brasseur, 2020). Similarly, in New  
282 York, stay-at-home orders and social restrictions were rigorously enforced by late March 2020  
283 (Tzortziou et al., 2022). These interventions contributed to substantial reductions in trace metal  
284 emissions, thereby mitigating associated health damages. In contrast, notable increases in health  
285 burden were observed in Northeast China, the southern coastal regions of China, and certain  
286 scattered areas in the Middle East. The rise in health risks linked to trace metal exposure in Northeast  
287 China and southern coastal regions may be associated with pollution aggravation during the  
288 COVID-19 period (Huang et al., 2021). Previous studies have highlighted that these regions  
289 experienced persistent air pollution or increased metal concentrations during the lockdown,



290 primarily due to unfavorable meteorological conditions (Li et al., 2023b). For instance, the  
291 prevalence of stable weather patterns, such as reduced wind speeds and atmospheric stagnation,  
292 significantly elevated ambient trace metal concentrations (McClymont and Hu, 2021; Şahin, 2020).

293 In our study, the global regions were categorized into eight major domains. During the COVID-  
294 19 period, China and India demonstrated the highest health benefits resulting from reductions in  
295 trace metal emissions. Additionally, Europe, NA, and Russia also exhibited significant health  
296 benefits. In contrast, SA, SS, and Australia showed the lowest health benefits, with some areas even  
297 experiencing an increase in health burden following the COVID-19 outbreak. The ambient trace  
298 metals in SA, SS, and Australia appeared to be less sensitive to lockdown measures due to relatively  
299 low baseline trace metal exposures in these regions. Combined the response of trace metal  
300 concentration to emission reduction in five scenarios, we also demonstrated that trace metal  
301 emission reductions are most effective in mitigating health damages in regions with high baseline  
302 exposures, such as China and India. Future efforts to target emission reductions in these regions  
303 could yield substantial public health benefits.

#### 304 **4 Conclusions and implications**

305 The global lockdowns during the COVID-19 pandemic significantly reduced anthropogenic  
306 emissions, yet the response of ambient trace metal concentrations to these control measures remains  
307 insufficiently understood. In this study, we developed an updated global trace metal emission  
308 inventory and employed a chemical transport model to predict the concentrations of nine trace  
309 metals for January-April in 2019 and 2020. Our results revealed that the response characteristics of  
310 trace metals to lockdown measures varied substantially. Global average concentrations of ambient  
311 As, Cd, Cr, Cu, Mn, Ni, and V decreased by 5%, 1%, 1%, 4%, 5%, 7%, and 7%, respectively,  
312 following the COVID-19 outbreak. However, global average concentrations of particulate Pb (1%)  
313 and Zn (2%) showed slight increases during the same period. This trend can be attributed to coal  
314 combustion and non-ferrous smelting industries, which are critical sectors for meeting residential  
315 needs and thus less responsive to emission control measures. Significant spatial variations in trace  
316 metal responses to lockdown measures were also observed. For instance, lockdown interventions  
317 were more effective in reducing trace metal pollution in India, Europe, and NA compared to other  
318 regions. Moreover, the trace metal concentrations show the linear response to emission reduction,



319 and thus the prioritizing emission reductions in heavily polluted areas (e.g., China, India, WE, and  
320 NA) yields higher marginal benefits and greater public health gains. From a health burden  
321 perspective, controlling emissions of Pb and As emerged as the most effective strategies for  
322 mitigating carcinogenic and non-carcinogenic risks, respectively. Both elements are primarily  
323 associated with fossil fuel combustion, particularly coal burning. However, the persistent increase  
324 in energy consumption poses a challenge to achieving meaningful reductions in Pb and As emissions.  
325 In the future, it will be essential to implement additional control measures to curb Pb and As  
326 emissions during coal combustion processes, thereby maximizing health benefits and reducing  
327 environmental risks.

328 It is important to acknowledge several limitations in our study. Firstly, the simulated  
329 concentrations of hazardous trace metals in SA and SS might contain uncertainties due to the  
330 scarcity of ground-level observations in these regions. This lack of observational data makes it  
331 challenging to accurately evaluate the predictive reliability of ambient trace metal concentrations in  
332 these areas. Consequently, these uncertainties may also propagate to the health risk assessments. To  
333 address this, it is crucial to establish more monitoring sites for ambient trace metals in SA and SS.  
334 Secondly, the health risk assessment in this study was based solely on trace metal concentrations,  
335 without considering population exposure. In reality, health impacts are closely tied to population  
336 size and distribution. Future research should prioritize the development of more accurate  
337 methodologies that incorporate population exposure to better assess the health impacts of ambient  
338 trace metal exposure.

### 339 **Acknowledgements**

340 This work was supported by the Opening Project of Shanghai Key Laboratory of Atmospheric  
341 Particle Pollution and Prevention (LAP3) [grant numbers FDLAP24002]; Academic Mentorship for  
342 Scientific Research Cadre Project [grant numbers AMSCP-24-05-03].

### 343 **Author Contributions**

344 Conceptualization: RL, Data Curation: WS and XL, Formal analysis: RL.

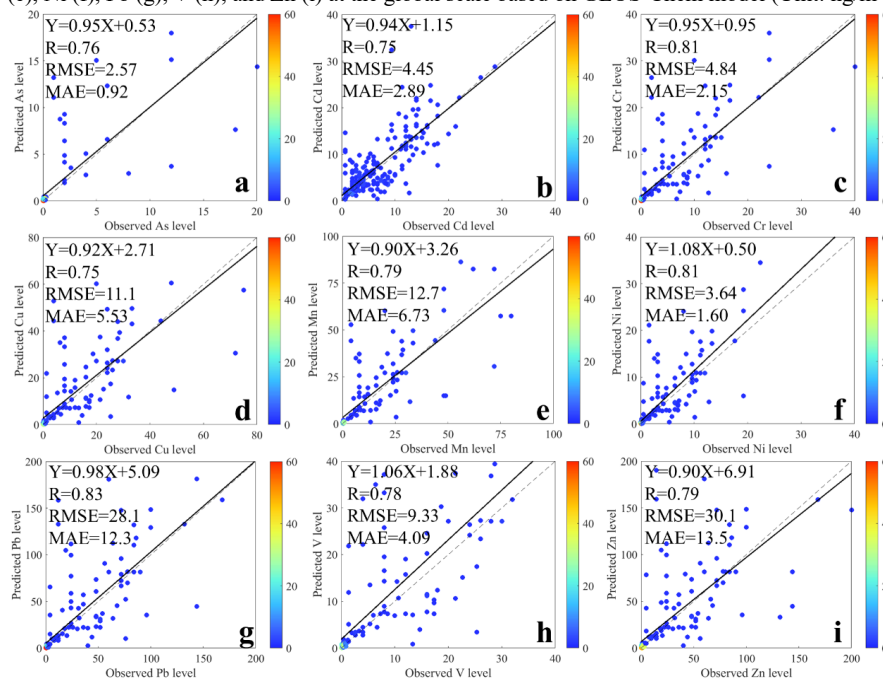
### 345 **Competing interests**

346 The contact author has declared that none of the authors has any competing interests.

347

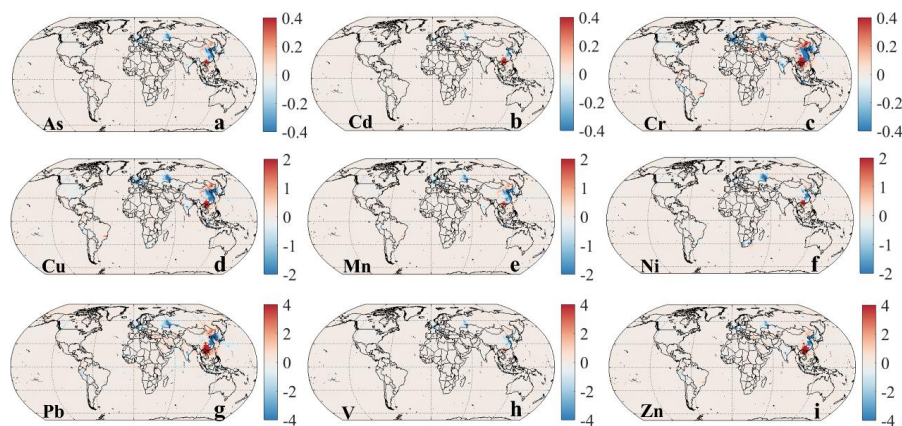


**Figure 1** The predictive accuracy of nine trace metals including As (a), Cd (b), Cr (c), Cu (d), Mn (e), Ni (f), Pb (g), V (h), and Zn (i) at the global scale based on GEOS-Chem model (Unit: ng/m<sup>3</sup>).



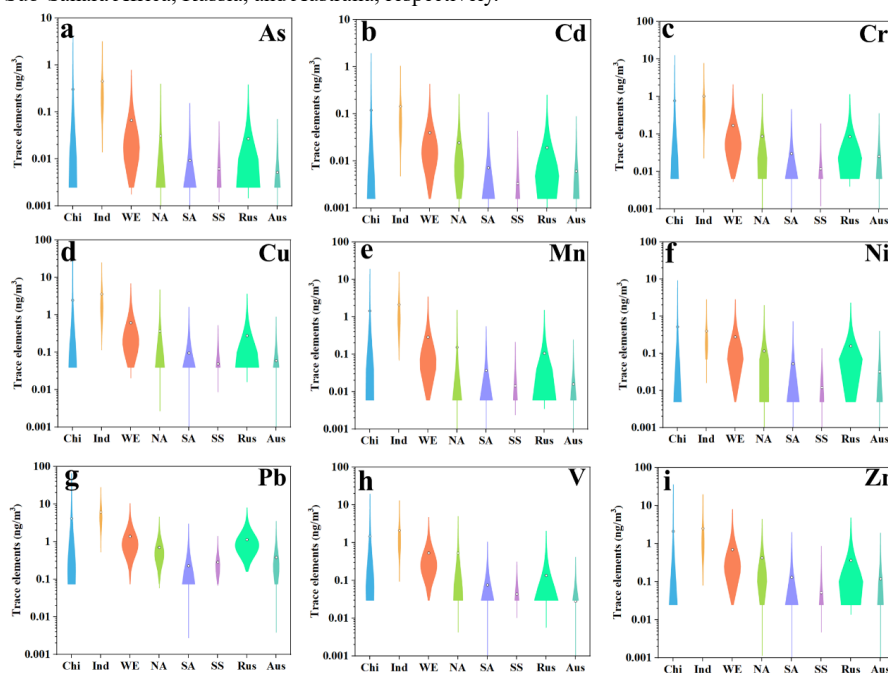


**Figure 2** The global difference of trace element concentrations between January-April in 2019 and 2020. Nine trace elements including As (a), Cd (b), Cr (c), Cu (d), Mn (e), Ni (f), Pb (g), V (h), and Zn (i) were selected to analyze the annual variations (Unit: ng/m<sup>3</sup>).



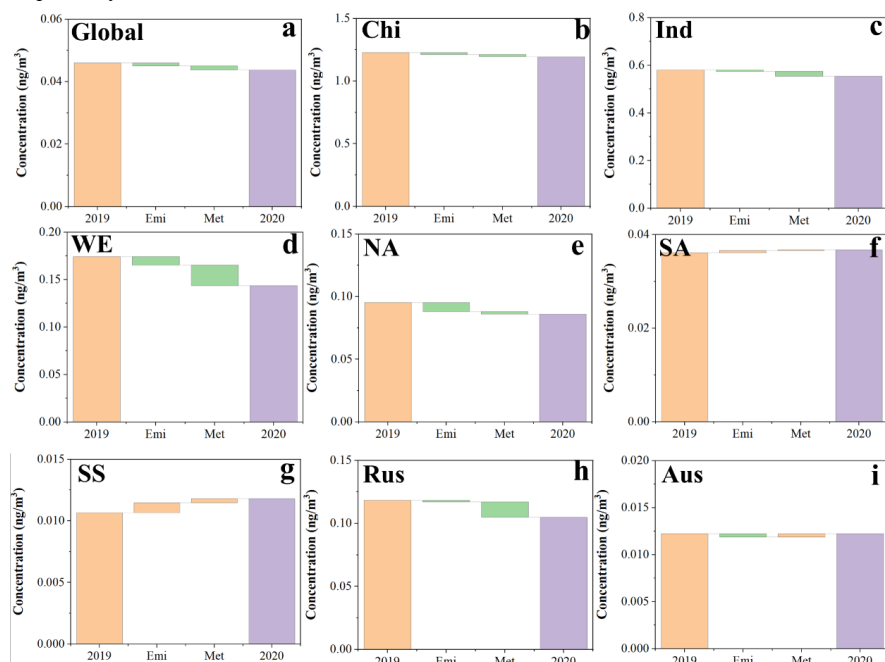


**Figure 3** The violin graphs of nine trace elements including As (a), Cd (b), Cr (c), Cu (d), Mn (e), Ni (f), Pb (g), V (h), and Zn (i) in eight major regions during January-April in 2020. Chi, Ind, WE, NA, SA, SS, Rus, and Aus represent China, India, Western Europe, North America, South America, Sub-Sahara Africa, Russia, and Australia, respectively.





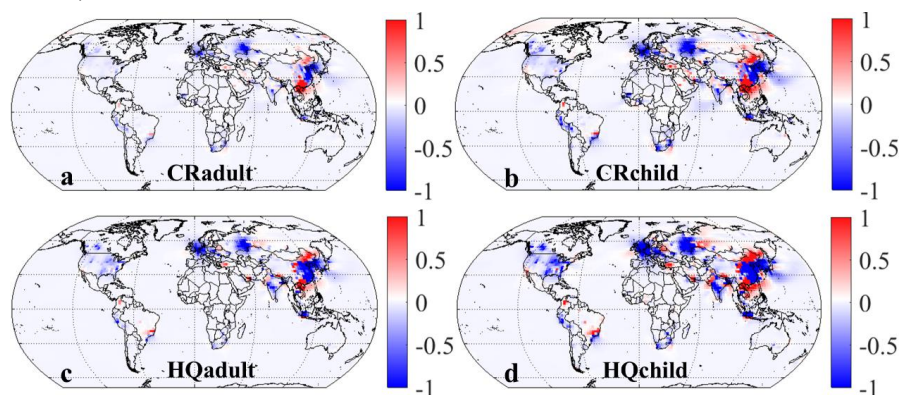
**Figure 4** The emission and meteorological contributions to ambient As concentrations during 2019-2020 at global and eight major regions. Chi, Ind, WE, NA, SA, SS, Rus, and Aus represent China, India, Western Europe, North America, South America, Sub-Sahara Africa, Russia, and Australia, respectively.







**Figure 5** The total CR and HQ differences of adults and children for all of the nine hazardous trace metals during January-April in 2019 and 2020 (the minus of CR and HQ values in 2020 and those in 2019).





## References

- Al-Sulaiti, M.M., Soubra, L., Al-Ghouti, M.A. (2022) The causes and effects of mercury and methylmercury contamination in the marine environment: A review. *Current Pollution Reports* 8, 249-272.
- Bai, X., Tian, H., Zhu, C., Luo, L., Hao, Y., Liu, S., Guo, Z., Lv, Y., Chen, D., Chu, B. (2023) Present knowledge and future perspectives of atmospheric emission inventories of toxic trace elements: a critical review. *Environmental science & technology* 57, 1551-1567.
- Chang, Y., Huang, R.J., Ge, X., Huang, X., Hu, J., Duan, Y., Zou, Z., Liu, X., Lehmann, M.F. (2020) Puzzling haze events in China during the coronavirus (COVID-19) shutdown. *Geophysical Research Letters* 47, e2020GL088533.
- Cheng, K., Wang, Y., Tian, H., Gao, X., Zhang, Y., Wu, X., Zhu, C., Gao, J. (2015) Atmospheric emission characteristics and control policies of five precedent-controlled toxic heavy metals from anthropogenic sources in China. *Environmental science & technology* 49, 1206-1214.
- Das, S., Prospero, J.M., Chellam, S. (2023) Quantifying international and interstate contributions to primary ambient PM<sub>2.5</sub> and PM<sub>10</sub> in a complex metropolitan atmosphere. *Atmospheric Environment* 292, 119415.
- Doumbia, T., Granier, C., Elguindi, N., Bouarar, I., Darras, S., Brasseur, G., Gaubert, B., Liu, Y., Shi, X., Stavrou, T. (2021) Changes in global air pollutant emissions during the COVID-19 pandemic: a dataset for atmospheric modeling. *Earth System Science Data* 13, 4191-4206.
- Farahani, V.J., Soleimani, E., Pirhadi, M., Sioutas, C. (2021) Long-term trends in concentrations and sources of PM<sub>2.5</sub>-bound metals and elements in central Los Angeles. *Atmospheric Environment* 253, 118361.
- Fonseca, E.M.d., Natrass, N., Lazaro, L.L.B., Bastos, F.I. (2021) Political discourse, denialism and leadership failure in Brazil's response to COVID-19. *Global public health* 16, 1251-1266.
- Guo, F., Tang, M., Wang, X., Yu, Z., Wei, F., Zhang, X., Jin, M., Wang, J., Xu, D., Chen, Z. (2022) Characteristics, sources, and health risks of trace metals in PM<sub>2.5</sub>. *Atmospheric Environment* 289, 119314.
- Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W. (2020) Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China. *National Science Review*.
- Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W. (2021) Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China. *National Science Review* 8, nwaa137.
- Huber, M., Welker, A., Helmreich, B. (2016) Critical review of heavy metal pollution of traffic area runoff: Occurrence, influencing factors, and partitioning. *Science of the Total Environment* 541, 895-919.
- Jia, M., Evangelou, N., Eckhardt, S., Huang, X., Gao, J., Ding, A., Stohl, A. (2021) Black carbon emission reduction due to COVID-19 lockdown in China. *Geophysical Research Letters* 48, e2021GL093243.
- Kan, X., Dong, Y., Feng, L., Zhou, M., Hou, H. (2021) Contamination and health risk assessment of heavy metals in China's lead-zinc mine tailings: A meta-analysis. *Chemosphere* 267, 128909.
- Keller, C.A., Evans, M.J., Knowland, K.E., Hasenkopf, C.A., Modekurty, S., Lucchesi, R.A., Oda, T., Franca, B.B., Mandarino, F.C., Díaz Suárez, M.V. (2021) Global impact of COVID-19 restrictions on the surface concentrations of nitrogen dioxide and ozone. *Atmospheric Chemistry and Physics*



- 21, 3555-3592.
- La Colla, N.S., Botté, S.E., Marcovecchio, J.E. (2021) Atmospheric particulate pollution in South American megacities. *Environmental reviews* 29, 415-429.
- Li, H., Yao, J., Sunahara, G., Min, N., Li, C., Duran, R. (2023a) Quantifying ecological and human health risks of metal (loid)s pollution from non-ferrous metal mining and smelting activities in Southwest China. *Science of the Total Environment* 873, 162364.
- Li, R., Peng, M., Zhao, W., Wang, G., Hao, J. (2022) Measurement Report: Rapid changes of chemical characteristics and health risks for high time-resolved trace elements in PM<sub>2.5</sub> in a typical industrial city response to stringent clean air actions. *EGUsphere* 2022, 1-37.
- Li, R., Zhang, L., Gao, Y., Wang, G. (2023b) Different Response Mechanisms of N-Bearing Components to Emission Reduction Across China During COVID-19 Lockdown Period. *Journal of Geophysical Research: Atmospheres* 128, e2023JD039496.
- Li, R., Zhao, Y., Fu, H., Chen, J., Peng, M., Wang, C. (2021) Substantial changes in gaseous pollutants and chemical compositions in fine particles in the North China Plain during the COVID-19 lockdown period: anthropogenic vs. meteorological influences. *Atmospheric Chemistry and Physics* 21, 8677-8692.
- Liu, H., Jacob, D.J., Bey, I., Yantosca, R.M. (2001) Constraints from <sup>210</sup>Pb and <sup>7</sup>Be on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields. *Journal of Geophysical Research: Atmospheres* 106, 12109-12128.
- Liu, S., Tian, H., Bai, X., Zhu, C., Wu, B., Luo, L., Hao, Y., Liu, W., Lin, S., Zhao, S. (2021) Significant but spatiotemporal-heterogeneous health risks caused by airborne exposure to multiple toxic trace elements in China. *Environmental science & technology* 55, 12818-12830.
- Loomis, D., Guha, N., Hall, A.L., Straif, K. (2018) Identifying occupational carcinogens: an update from the IARC Monographs. *Occupational and environmental medicine* 75, 593-603.
- Mao, J., Jacob, D.J., Evans, M., Olson, J., Ren, X., Brune, W., St Clair, J., Crounse, J., Spencer, K., Beaver, M. (2010) Chemistry of hydrogen oxide radicals (HO<sub>x</sub>) in the Arctic troposphere in spring. *Atmospheric Chemistry and Physics* 10, 5823-5838.
- Marufi, N., Conti, G.O., Ahmadinejad, P., Ferrante, M., Mohammadi, A.A. (2024) Carcinogenic and non-carcinogenic human health risk assessments of heavy metals contamination in drinking water supplies in Iran: a systematic review. *Reviews on Environmental Health* 39, 91-100.
- McClymont, H., Hu, W. (2021) Weather variability and COVID-19 transmission: a review of recent research. *International Journal of Environmental Research and Public Health* 18, 396.
- Meo, S.A., Abukhalaf, A.A., Alomar, A.A., AlMutairi, F.J., Usmani, A.M., Klonoff, D.C. (2020) Impact of lockdown on COVID-19 prevalence and mortality during 2020 pandemic: observational analysis of 27 countries. *European journal of medical research* 25, 1-7.
- Nirmalkar, J., Haswani, D., Singh, A., Kumar, S., Raman, R.S. (2021) Concentrations, transport characteristics, and health risks of PM<sub>2.5</sub>-bound trace elements over a national park in central India. *Journal of Environmental Management* 293, 112904.
- Onyeaka, H., Anumudu, C.K., Al-Sharify, Z.T., Egele-Godswill, E., Mbaegbu, P. (2021) COVID-19 pandemic: A review of the global lockdown and its far-reaching effects. *Science progress* 104, 00368504211019854.
- Pacyna, J.M., Pacyna, E.G. (2001) An assessment of global and regional emissions of trace metals to the atmosphere from anthropogenic sources worldwide. *Environmental reviews* 9, 269-298.
- Pan, Y., Wang, Y. (2015) Atmospheric wet and dry deposition of trace elements at 10 sites in Northern



- China. *Atmospheric Chemistry and Physics* 15, 951-972.
- Park, R.J., Jacob, D.J., Field, B.D., Yantosca, R.M., Chin, M. (2004) Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy. *Journal of Geophysical Research: Atmospheres* 109.
- Pearce, N., Blair, A., Vineis, P., Ahrens, W., Andersen, A., Anto, J.M., Armstrong, B.K., Baccarelli, A.A., Beland, F.A., Berrington, A. (2015) IARC monographs: 40 years of evaluating carcinogenic hazards to humans. *Environmental health perspectives* 123, 507-514.
- Qiu, Y., Ma, Z., Li, K., Lin, W., Tang, Y., Dong, F., Liao, H. (2020) Markedly enhanced levels of peroxyacetyl nitrate (PAN) during COVID-19 in Beijing. *Geophysical Research Letters* 47, e2020GL089623.
- Saadat, S., Rawtani, D., Hussain, C.M. (2020) Environmental perspective of COVID-19. *Science of the Total Environment* 728, 138870.
- Şahin, M. (2020) Impact of weather on COVID-19 pandemic in Turkey. *Science of the Total Environment* 728, 138810.
- Sharma, A.K., Sharma, M., Sharma, A.K., Sharma, M. (2023) Mapping the impact of environmental pollutants on human health and environment: A systematic review and meta-analysis. *Journal of Geochemical Exploration*, 107325.
- Shi, X., Brasseur, G.P. (2020) The Response in Air Quality to the Reduction of Chinese Economic Activities during the COVID-19 Outbreak. *Geophysical Research Letters*, e2020GL088070.
- Smith, L.V., Tarui, N., Yamagata, T. (2021) Assessing the impact of COVID-19 on global fossil fuel consumption and CO<sub>2</sub> emissions. *Energy economics* 97, 105170.
- Streets, D.G., Devane, M.K., Lu, Z., Bond, T.C., Sunderland, E.M., Jacob, D.J. (2011) All-time releases of mercury to the atmosphere from human activities. *Environmental science & technology* 45, 10485-10491.
- Tikadar, K.K., Jahan, F., Mia, R., Rahman, M.Z., Sultana, M.A., Islam, S., Kunda, M. (2024) Assessing the potential ecological and human health risks of trace metal pollution in surface water, sediment, and commercially valuable fish species in the Pashur River, Bangladesh. *Environmental Monitoring and Assessment* 196, 1042.
- Tørseth, K., Aas, W., Breivik, K., Fjærraa, A.M., Fiebig, M., Hjellbrekke, A.G., Lund Myhre, C., Solberg, S., Yttri, K.E., 2012. Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972-2009. *Atmos. Chem. Phys.* 12, 544-5481.
- Tzortziou, M., Kwong, C.F., Goldberg, D., Schiferl, L., Commane, R., Abuhassan, N., Szykman, J.J., Valin, L.C. (2022) Declines and peaks in NO<sub>2</sub> pollution during the multiple waves of the COVID-19 pandemic in the New York metropolitan area. *Atmospheric Chemistry and Physics* 22, 2399-2417.
- Venter, Z.S., Aunan, K., Chowdhury, S., Lelieveld, J. (2020) COVID-19 lockdowns cause global air pollution declines. *Proceedings of the National Academy of Sciences* 117, 18984-18990.
- Wesely, M. (2007) Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. *Atmospheric Environment* 41, 52-63.
- Wu, Y., Lin, S., Tian, H., Zhang, K., Wang, Y., Sun, B., Liu, X., Liu, K., Xue, Y., Hao, J. (2020) A quantitative assessment of atmospheric emissions and spatial distribution of trace elements from natural sources in China. *Environmental pollution* 259, 113918.
- Xing, X., Xiong, Y., Yang, R., Wang, R., Wang, W., Kan, H., Lu, T., Li, D., Cao, J., Peñuelas, J. (2021)



- Predicting the effect of confinement on the COVID-19 spread using machine learning enriched with satellite air pollution observations. *Proceedings of the National Academy of Sciences* 118, e2109098118.
- Yu, S., Zhu, Y.-g., Li, X.-d. (2012) Trace metal contamination in urban soils of China. *Science of the Total Environment* 421, 17-30.
- Zhang, H., Zhang, F., Song, J., Tan, M.L., Johnson, V.C. (2021) Pollutant source, ecological and human health risks assessment of heavy metals in soils from coal mining areas in Xinjiang, China. *Environmental Research* 202, 111702.
- Zhang, L., Gao, Y., Wu, S., Zhang, S., Smith, K.R., Yao, X., Gao, H. (2020) Global impact of atmospheric arsenic on health risk: 2005 to 2015. *Proceedings of the National Academy of Sciences* 117, 13975-13982.
- Zhou, J., Tian, H., Zhu, C., Hao, J., Gao, J., Wang, Y., Xue, Y., Hua, S., Wang, K. (2015) Future trends of global atmospheric antimony emissions from anthropogenic activities until 2050. *Atmospheric Environment* 120, 385-392.
- Zhu, C., Tian, H., Hao, Y., Gao, J.J., Hao, J., Wang, Y., Liu, H.J. (2018) A high-resolution emission inventory of anthropogenic trace elements in Beijing-Tianjin-Hebei (BTH) region of China. *Atmospheric Environment* 191, 452-462.
- Zhu, C., Tian, H., Hao, J. (2020) Global anthropogenic atmospheric emission inventory of twelve typical hazardous trace elements, 1995–2012. *Atmospheric Environment* 220, 117061.