Returning impact of long-range PM2.5 transport over East Asia in 2021 after Chinese economic recovery from the COVID-19 pandemic

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Abstract

Changes in the aerosol composition of sulfate (SO$_{4}^{2-}$) and nitrate (NO$_{3}^{-}$) from 2012 to 2019 have been captured as a paradigm shift in the region downwind of China. Specifically, SO$_{4}^{2-}$ dramatically decreased and NO$_{3}^{-}$ dramatically increased over downwind locations such as western Japan due to the faster reduction of SO$_2$ emissions than NO$_x$ emissions and the almost constant trend of NH$_3$ emissions from China. Emissions from China sharply decreased during COVID-19 lockdowns in February–March 2020, after which China’s economic situation seemed to recover going into 2021. Given this substantial change in Chinese emissions, it is necessary to clarify the impact of long-range PM$_{2.5}$ transport over East Asia. In this study, ground-based aerosol compositions observed at three sites in western Japan were analysed. The concentrations of PM$_{2.5}$, SO$_{4}^{2-}$ and NO$_{3}^{-}$ decreased in 2020 (during COVID-19) compared with 2018–2019 (before COVID-19). In 2021 (after COVID-19), PM$_{2.5}$ and NO$_{3}^{-}$ increased and SO$_{4}^{2-}$ was unchanged. This suggests the returning impact of long-range PM$_{2.5}$ transport in 2021. From numerical simulations, the status of Chinese emissions during COVID-19 did not explain this returning impact in 2021. This study shows that the status of Chinese emissions in 2021 was close to that before COVID-19.

Introduction

Before the outbreak of COVID-19, strong emission regulations in China had led to a rapid decrease in PM$_{2.5}$ concentrations from 2013$^{1-3}$, and this improvement in Chinese PM$_{2.5}$ pollution was associated with improved air quality in terms of PM$_{2.5}$ over the region downwind of China in 2010–2019$^{4}$. Moreover, nitrate aerosol (NO$_{3}^{-}$) became an important component of PM$_{2.5}$ by replacing sulfate aerosol (SO$_{4}^{2-}$), which had previously dominated aerosol pollution over East Asia$^{5,6}$. This feature is recognised as a paradigm shift in PM$_{2.5}$ composition. Such variations with increasing importance of NO$_{3}^{-}$ were also found from long-term analyses of the precipitation chemistry over East Asia$^{7,8}$. These changes were due to the different reduction rates of NO$_x$ and SO$_2$ emissions with the faster reduction in SO$_2$ emission and the steadily high rate of NH$_3$ emission in China. This trend was expected to continue.

However, unexpected changes in air quality occurred because of the COVID-19 outbreak in December 2019 in the city of Wuhan in Hubei, China$^{9}$. In response to the pandemic, lockdowns were imposed on many cities in China during February and March 2020, and these measures limited human activities such as travel and economic activity$^{10,11}$. Accordingly, strong reductions in anthropogenic emissions were observed$^{12,13}$, and subsequent improvements in air pollution were reported over China$^{14,15}$. Because the long-range transport (i.e. trans-boundary transport) of air pollutants in East Asia is an important environmental issue$^{12,16,17}$, the reduction in anthropogenic emissions in China during COVID-19 will further impact the changes in air pollution over not only China but also downwind regions such as the Korean Peninsula and Japan. This leads to another question: How is the situation after COVID-19? Based on China’s estimated GDP, its economy has recovered since the 2020 lockdowns and is projected to keep
growing. Unlike the OECD countries, China achieved positive GDP growth in 2020, and this economic recovery likely led to a rebound in anthropogenic emissions. Against this background, it is worth investigating and clarifying, from the perspective of the downwind region, the changes in long-range PM$_{2.5}$ transport that resulted from the substantial emission changes that happened before, during and after COVID-19 in China. In the present study, we analysed the changes in aerosol components over western Japan based on a combination of ground-based and satellite observations and sensitivity experiments in numerical modeling simulations.

**Results**

We analysed aerosol compositions for February and March in each year from 2018 to 2021. For simplicity in the following discussion, 2018, for example, stands for the average in February and March in 2018. These two months corresponded to COVID-19 lockdown periods in China in 2020 and so are considered as the most suitable period for evaluating the change in PM$_{2.5}$ pollution before, during and after COVID-19. Herein, we refer to the two-year average for 2018 and 2019 as before COVID-19, 2020 as during COVID-19, and 2021 as after COVID-19. Figure 1 shows the NO$_2$ column over East Asia as measured by the Ozone Monitoring Instrument (OMI) (see Methods) before, during, and after COVID-19. A high NO$_2$ column was found over north-eastern China before COVID-19 (Fig. 1a). Compared with before COVID-19 status, the NO$_2$ column decreased dramatically during COVID-19 over the whole of eastern China (Fig. 1b). Then, although some parts showed decreases, almost all parts of China showed an increased NO$_2$ column after COVID-19 compared to during it (Fig. 1c). These satellite results suggest substantial decreases in emissions over China in 2020 and then increased emissions along with the economic recovery in 2021.

Figure 2 shows the observational results of mean, 75 percentile (3Q), median, and 25 percentile (1Q) for changes in PM$_{2.5}$ and its composition at three remote sites in Goto, Tsushima and Oki located in western Japan (see the lower-right corner of Fig. 1c) from 2018 to 2021. At each of these three sites, an aerosol chemical speciation analyzer (ACSA) measured the PM$_{2.5}$, SO$_4^{2-}$ and NO$_3^-$ concentrations (see Methods). Because NO$_3^-$ concentration had high concentration peak in short-time and generally close to zero, the value of mean are sometimes higher than the value of 3Q. In addition to the concentration analyses in Fig. 2, changes in the concentration of PM$_{2.5}$ and its composition (denoted as $C$) during and after COVID-19 are further illustrated in Fig. 3 based on the following equations:

\[
\text{Change in } C_{\text{during COVID-19}} [\%] = \left( \frac{C_{\text{during COVID-19}} - C_{\text{before COVID-19}}}{C_{\text{before COVID-19}}} \right) \times 100, (1)
\]

\[
\text{Change in } C_{\text{after COVID-19}} [\%] = \left( \frac{C_{\text{after COVID-19}} - C_{\text{during COVID-19}}}{C_{\text{during COVID-19}}} \right) \times 100. (2)
\]
The observational results (Figs. 2 and 3) show that the PM$_{2.5}$, SO$_4^{2-}$ and NO$_3^-$ concentrations decreased substantially during compared with before COVID-19. The SO$_4^{2-}$ concentration decreased gradually from 2018 to 2020, whereas the NO$_3^-$ concentration increased from 2018 to 2019 and then decreased from 2019 to 2020. The increase in NO$_3^-$ concentration in 2019 found at Goto and Oki was consistent with our previous study that reported the change in aerosol composition as a paradigm shift$^4$. In 2021, increases in PM$_{2.5}$ and NO$_3^-$ and almost the same level of SO$_4^{2-}$ have been measured. Considering the expected economic recovery suggested from the NO$_2$ column (Fig. 1c), the returning impact of long-range transport can be assumed. However, because PM$_{2.5}$ concentration and composition can also be affected by the meteorological field and emission sources other than anthropogenic sources, further discussion is required, and we conducted modeling simulations to clarify this point.

**Discussion**

To identify a potential reason for the variation in air pollutants found in 2021, we conducted numerical simulations for 2018–2021. The experiments are described in Table 1 (see Methods for details about the modeling configurations). In the names of these experiments, ‘M’ and ‘E’ denote meteorology and Chinese anthropogenic emissions, respectively (e.g. M18E18 indicates the meteorological field for 2018 and the emission inventory for 2018). From 2018 to 2020, the meteorological field and available emissions corresponding to the simulation periods were used. Through comparison with ACSA observations for 2018–2020 (Figs. 2 and 3), we confirmed that the model generally captured the observed variations of PM$_{2.5}$, SO$_4^{2-}$ and NO$_3^-$ concentrations. Statistical analyses were conducted to evaluate the modeling performance based on the correlation coefficient ($R$), the normalised mean bias ($NMB$) and the normalised mean error ($NME$), and the modeling performance was judged based on metrics from a review of modeling applications (see Methods for definitions). The results of the statistical analyses are given in Table 2. For PM$_{2.5}$, the model showed moderate correlation with observations but tended to underestimate the observed concentration; in some cases, the criteria for acceptable model performance were judged as not having been met. This underestimation stemmed mainly from the underestimation of organic aerosol in comparison with a model intercomparison study$^{19}$. For SO$_4^{2-}$ and NO$_3^-$, the model also tended to underestimate the observed concentrations, but in most cases it was judged to have performed well within the criteria. The model's underestimation for SO$_4^{2-}$ was due to the insufficient oxidation process in the current modeling$^{20}$. See our previous work for several possible reasons of the model's underestimation for NO$_3^-$ $^{21,22}$. 


Table 1
Description of present modeling experiments (‘M’ and ‘E’ denote meteorology and Chinese anthropogenic emissions, respectively).

| Modeling experiment | Meteorology | Anthropogenic emissions from China | Ship emissions | Natural emissions (biomass burning, biogenic and volcanoes) | Purpose |
|---------------------|-------------|-----------------------------------|----------------|-------------------------------------------------|---------|
| M18E18              | 2018        | 2018                              | 2010           | 2018                                            | Simulation using meteorology and emissions for 2018 |
| M19E19              | 2019        | 2019                              | 2010           | 2019                                            | Simulation using meteorology and emissions for 2019 |
| M20E20              | 2020        | 2020                              | 2020           | 2020                                            | Simulation using meteorology and emissions for 2020 |
| M21E20              | 2021        | 2020                              | 2020           | 2021                                            | Simulation using meteorology and fixed Chinese emissions in 2020 for 2021 assuming the emission levels during COVID-19 |
| M21E19              | 2021        | 2019                              | 2020           | 2021                                            | Simulation using meteorology and fixed Chinese emissions in 2019 for 2021 assuming the emission levels before COVID-19 |
Table 2
Statistical analyses of modeling performance.

| Location | Pollutant | Metrics | Modeling experiment |
|----------|-----------|---------|---------------------|
|          |           |         | M18E18 | M19E19 | M20E20 | M21E20 | M21E19 |
| Goto     | PM$_{2.5}$ | $R$     | 0.83** | 0.81** | 0.76** | —      | —      |
|          |           | $NMB$ (%) | −37.1  | −34.7  | −22.1* | —      | —      |
|          |           | $NME$ (%) | 40.5*  | 40.3*  | 44.4*  | —      | —      |
|          | SO$_4^{2-}$ | $R$     | 0.79** | 0.76** | 0.48*  | —      | —      |
|          |           | $NMB$ (%) | −25.6* | −17.4* | −3.2** | —      | —      |
|          |           | $NME$ (%) | 38.4*  | 37.3*  | 59.3   | —      | —      |
|          | NO$_3^-$  | $R$     | 0.57   | 0.60   | 0.83   | —      | —      |
|          |           | $NMB$ (%) | −51.2* | −37.2* | −32.8* | —      | —      |
|          |           | $NME$ (%) | 70.6*  | 66.8*  | 63.7** | —      | —      |
| Tsushima | PM$_{2.5}$ | $R$     | —      | —      | 0.71** | 0.59*  | 0.65*  |
|          |           | $NMB$ (%) | —      | —      | −22.6* | −56.6  | −46.5  |
|          |           | $NME$ (%) | —      | —      | 48.5*  | 57.8   | 49.6*  |
|          | SO$_4^{2-}$ | $R$     | —      | —      | 0.55*  | 0.66*  | 0.69*  |
|          |           | $NMB$ (%) | —      | —      | −2.1** | −26.1* | −10.3* |
|          |           | $NME$ (%) | —      | —      | 53.6   | 48.1*  | 45.9*  |
|          | NO$_3^-$  | $R$     | —      | —      | 0.81   | 0.65   | 0.65   |
|          |           | $NMB$ (%) | —      | —      | −22.9* | −63.6* | −36.5* |
|          |           | $NME$ (%) | —      | —      | 58.6*  | 73.0*  | 63.8** |
| Oki      | PM$_{2.5}$ | $R$     | 0.75** | 0.66*  | 0.48*  | 0.61*  | 0.62*  |

Notes: for PM$_{2.5}$ and SO$_4^{2-}$, the model performance goals (marked by **) are $R > 0.70$, $NMB < ±10\%$ and $NME < +35\%$, and the model performance criteria (marked by *) are $R > 0.4$, $NMB < ±30\%$ and $NME < +50\%$; for NO$_3^-$, the model performance goals (marked by **) are $NMB < ±15\%$ and $NME < +65\%$, and the model performance criteria (marked by *) are $NMB < ±65\%$ and $NME < +115\%$. Where M21E19 is superior to M21E20 in a statistical score, the value is given in bold font.
### Table

|          | NMB (%) | NME (%) | R   | NMB (%) | NME (%) |
|----------|---------|---------|-----|---------|---------|
| **SO₄²⁻** |         |         |     |         |         |
|          | -29.3*  | 39.1*   | 0.72** | -20.0*  | 42.6*   |
|          | -42.3   | 46.2*   | 0.71** | -22.7*  | 35.8*   |
|          | -37.1   | 54.0    | 0.55*  | -10.8*  | 40.8*   |
|          | -49.2   | 54.5    | 0.71** | -17.2*  | 38.1*   |
|          | -36.8   | 47.7*   | 0.69*  | -1.8**  | 40.1*   |
| **NO₃⁻** |         |         |     |         |         |
|          | -16.9*  | 76.4*   | 0.51  | -20.8*  | 70.1*   |
|          | -32.5*  | 74.8*   | 0.54  | -55.3*  | 74.8*   |
|          | -55.3*  | 75.2*   | 0.56  | -22.7*  | 74.8*   |

Notes: for PM$_{2.5}$ and SO$_{4}^{2−}$, the model performance goals (marked by **) are $R > 0.70$, $NMB < ±10\%$ and $NME < +35\%$, and the model performance criteria (marked by *) are $R > 0.4$, $NMB < ±30\%$ and $NME < +50\%$; for NO$_{3}^{−}$, the model performance goals (marked by **) are $NMB < ±15\%$ and $NME < +65\%$, and the model performance criteria (marked by *) are $NMB < ±65\%$ and $NME < +115\%$. Where M21E19 is superior to M21E20 in a statistical score, the value is given in bold font.

For 2021, two experiments were conducted because of the lack of emission information for that year (Table 1). In these experiments, anthropogenic emissions from China have been perturbed. One was M21E20, using the meteorological field for 2021 but the Chinese emissions for 2020 (as during COVID-19) with the natural emissions for 2021; the other was M21E19, also using the meteorological field for 2021 but now the Chinese emissions for 2019 (as before COVID-19) with the natural emissions for 2021. Based on these two experiments, the impact of supposing emission changes in 2021 can be estimated. From comparing the simulation results and the ACSA observations for Tsushima and Oki in 2021 (Fig. 2), the observations showed increases in PM$_{2.5}$ and NO$_{3}^{−}$ but almost none in SO$_{4}^{2−}$, whereas M21E20 showed decreases except for a slight increase in NO$_{3}^{−}$ at Oki. The results from M21E20 did not explain the variation observed in 2021, whereas M21E19 gave almost flat variations in PM$_{2.5}$ and SO$_{4}^{2−}$ and an increase in NO$_{3}^{−}$. The results from M21E19 were much closer to the observational results. The relative changes in the PM$_{2.5}$, SO$_{4}^{2−}$ and NO$_{3}^{−}$ concentrations after COVID-19 (Fig. 3) also clarified the differences in modeling performance for 2021. Experiment M21E20 showed negative changes after COVID-19 except for NO$_{3}^{−}$ at Oki, results that are not consistent with the observations, whereas M21E19 showed better agreement with the observations. These analyses show that M21E19 outperformed M21E20 in capturing the changes observed after COVID-19. In terms of statistical scores (Table 2), M21E19 also outperformed M21E20. These results clarify that the emission status in 2021 was not the same as during COVID-19 and was close to the status before COVID-19. Chinese economic activity and anthropogenic emissions recovered in 2021 and impacted the air quality in downwind regions through long-range PM$_{2.5}$ transport.
Note that experiment M21E20 gave negative changes after COVID-19. Based on the differences between M20E20 and M21E20 (i.e. the M21E20 results minus the M20E20 ones), the impact due to the meteorology and changes in natural emission in 2021 can be investigated. The model-estimated changes in the concentrations of PM$_{2.5}$, SO$_4$$_{2}^{2-}$ and NO$_3$$_{-}$ caused by meteorology and natural emissions are shown in Fig. 4a–c. The negative changes in PM$_{2.5}$, SO$_4$$_{2}^{2-}$ and NO$_3$$_{-}$ concentrations over the East China Sea to the Korean Peninsula and western Japan are shown clearly (Fig. 4a–c). Based on the analyses of changes in natural emissions, volcanoes distributed mainly over the western island of Kyushu$^{23,24}$ showed SO$_2$ emissions of 605 Gg/period in 2020 and 307 Gg/period in 2021. This decrease could partly explain the negative change in 2021 as found in SO$_4$$_{2}^{2-}$, but a negative change in SO$_4$$_{2}^{2-}$ will lead to a positive change in NO$_3$$_{-}$ as examined previously as a paradigm shift in aerosol composition. Therefore, the negative changes seen for PM$_{2.5}$, SO$_4$$_{2}^{2-}$ and NO$_3$$_{-}$ can be attributed to changes in the meteorological field. To clarify this point, several meteorological fields are shown in Fig. S1 as the difference between 2021 and 2020 (i.e. the M21 meteorological fields minus the M20 ones) overlaid with the observational results. The positive relative humidity over the East China Sea may enhance the aqueous-phase reaction of SO$_4$$_{2}^{2-}$ and did not relate to negative changes in PM$_{2.5}$ and SO$_4$$_{2}^{2-}$ (Fig. S1a). The change in precipitation was inhomogeneous over the East China Sea to western Japan (Fig. S1b). Lower planetary boundary layer (PBL) height found over oceans will lead to suppressed pollutants and did not explain the negative concentrations (Fig. S1c). The change in the 2-m temperature partly explains the NO$_3$$_{-}$ change as understood by thermodynamic NO$_3$$_{-}$ production. The positive and negative temperatures over western and eastern Japan (Fig. S1d) are generally consistent with the negative and positive changes in NO$_3$$_{-}$ concentration over these areas (Fig. 4c and Fig. S1d). The positive change in the 10-m wind speed could contribute to the negative changes in concentrations because of the faster transport of polluted air mass (Fig. S1e). The most plausible explanation comes from the change in the 10-m wind direction (Fig. S1f), which was positive over eastern China to the East China Sea and western Japan. Further detailed illustrations of wind field are shown in Fig. 4d and e. During winter, the seasonal wind pattern is generally in the northwest direction$^5$, and a northwest wind direction was simulated over the East China Sea to western Japan in 2020 (Fig. 4d); however, a north wind direction dominated in 2021 over western Japan (Fig. 4e). Therefore, a positive difference in wind direction with positive wind speed was calculated over western Japan (Fig. 4f and Figs. S1e and f). The results observed at Tsushima and Oki are also displayed (Fig. S2), and the observations at Tsushima show clearly the change in wind direction from NNW in 2020 to N in 2021. These findings suggest that the meteorological conditions in 2021—especially the wind direction—prevented long-range PM$_{2.5}$ transport. Nevertheless, the observations showed increases in PM$_{2.5}$ and NO$_3$$_{-}$ and almost no change in SO$_4$$_{2}^{2-}$ in 2021. This point supports the assertion that the changes in concentrations in 2021 cannot be explained without the returning impact of long-range PM$_{2.5}$ transport associated with the growth in Chinese anthropogenic emissions after COVID-19.

Finally, as an extension of the analysis in our previous study$^4$, the SO$_4$$_{2}^{2-}$ and NO$_3$$_{-}$ variations before, during and after COVID-19 are summarised in Fig. 5. The scatterplot of SO$_4$$_{2}^{2-}$/NO$_3$$_{-}$ at Goto from before
to during COVID-19 has an inverted V shape. In this figure, the variations of \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) reported as a paradigm shift in our previous study\(^4\) are overlaid (grey open circles with a linear fitting line). These observation data\(^4\) were measured using a quadruple-type aerosol chemical speciation monitor (Q-ACSM) for fine aerosols of PM\(_1\), whereas the present study analysed PM\(_{2.5}\) components using an ACSA. The variations of \( \text{SO}_4^{2-} \) decrease and \( \text{NO}_3^- \) increase were also found based on ACSA observations up to 2019, and the dramatic decreases in both \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) during COVID-19 were furthermore clarified in the present study. The modeling simulation also reproduced well this inverted V-shaped variation. At Tsushima, the variations from during COVID-19 to after COVID-19 were relatively small. From the observational results, \( \text{SO}_4^{2-} \) did not change but \( \text{NO}_3^- \) increased slightly, and the model results also show small variations. Note that experiment M21E20 (grey closed circles) gave deviations from these small variations in both \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \). These results also show that assuming the emission status from during COVID-19 for 2021 was not appropriate. For further improvement of the modeling for Tsushima, the updated emission status over the Republic of Korea should be used. At Oki, the scatterplot of \( \text{SO}_4^{2-}/\text{NO}_3^- \) from before to during COVID-19 also has an inverted V shape as found at Goto. The variation from during COVID-19 to after COVID-19 shows no change in \( \text{SO}_4^{2-} \) but an increase in \( \text{NO}_3^- \). Experiment M21E19 reproduced this variation, but M21E20 did not. These analyses of the scatterplot of \( \text{SO}_4^{2-}/\text{NO}_3^- \) show substantial decreases in both \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) during COVID-19 but suggest the return to the situation of aerosol composition change regarding the importance of \( \text{NO}_3^- \) after COVID-19. The status of aerosol composition change should be monitored continuously because the nitrogen burden over East Asia is an issue of concern both currently and for the future\(^{25,26}\). The atmospheric input of nitrogen via dry and wet deposition processes over East Asia has been estimated in previous studies, and its significance has been clarified\(^{27-30}\). Targeting the proper management of nitrogen, the nitrogen burden during and after COVID-19 should be clarified in future work.

**Methods**

**Observations**

To obtain the spatial distribution of the NO\(_2\) column, which can be regarded as a proxy for NO\(_x\) emissions\(^31\), space-borne OMI measurements using the level-3 daily global nitrogen dioxide product (OMNO2d version 3.0) gridded into 0.25°×0.25°\(^32\) was used. We analysed the tropospheric column with clouds screened under the condition of a cloud fraction of less than 30%.

For ground-based observation of PM\(_{2.5}\) and its components, ACSA observation datasets for 10 sites in Japan were available\(^33\). The Japanese Ministry of the Environment (MoE) created this observation network in April 2017, and two remote observation sites at Goto and Oki, located in western Japan, were analysed in the present study. The ACSA measured PM\(_{2.5}\) and the secondary inorganic components of \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) with an hourly temporal resolution; this high temporal resolution minimises the effect of
the volatilisation of NO$_3^-$ 34. Observations at the Goto site were unavailable from the autumn of 2020 onwards because of a typhoon; to compensate for these missing observations at Goto, observations at the Tsushima site were used for 2020 and 2021. The locations of the three sites at Goto, Tsushima and Oki are shown in Fig. 1c.

The lockdown in Wuhan began on 23 January 2020 and lasted until 8 April 2020, and those in other Chinese cities occurred within this period 35. In Japan, a state of emergency was proclaimed for seven prefectures including Tokyo on 7 April 2020, and this was extended nationwide on 16 April 2020; it ended on 14 May 2020 for 39 prefectures except for mega-cities such Osaka and Tokyo, and on 25 May 2020 for all prefectures in Japan 36. As also seen from the Stringency Index released from One World Data 37, the countermeasures against COVID-19 were the strictest in China from late January to the end of March, whereas other countries in East Asia had relatively loose measures in place during this period. Given these differences between China and Japan, the period analysed in the present study was chosen as February to March to focus on the variation of long-range PM$_{2.5}$ transport according to Chinese anthropogenic emissions.

### Numerical modeling simulations

The numerical modeling simulations were performed using the regional chemical transport model of the Comprehensive Air quality Model with eXtensions (CAMx) version 6.4 38. The simulation domain covering East Asia was configured by 215×120 grid points with 36-km horizontal resolution, centred at 116.2°E and 37.8°N on a Lambert conformal projection. The vertical resolution was 44 non-uniformly spaced layers from the surface to 50 hPa 30. The meteorological fields to drive CAMx were calculated by the Weather Research and Forecasting (WRF) model version 4.1.1 39. Gas-phase and aerosol chemistry were respectively adopted for SAPRC07TC 40,41 and the CF scheme in CAMx. Based on previous findings 42,43, the NO$_2$ aqueous-phase reaction in the SO$_4^{2-}$ oxidation process was added. In addition, to enhance the production of SO$_4^{2-}$ during winter haze events, the heterogeneous reaction of SO$_2$ on aerosol surfaces was introduced following the work by Zheng 44. The initial and lateral boundary conditions were prepared from winter-averaged values of the extended CMAQ model over the northern hemisphere 45.

In the present study, five modeling experiments were conducted, and the simulation setups are described in Table 1. In the names of these experiments, ‘M’ and ‘E’ stand for meteorology and anthropogenic emissions, respectively. For each year, the corresponding meteorological field generated by WRF was used. The emissions inventories were prepared as follows. Anthropogenic emissions except those in Japan and China were based on Hemispheric Transport of Air Pollution (HTAP) version 2.2 46. For Japan, the latest available domestic emission inventory for 2015 developed by the MoE was used. For China, the emission projections were based on the Multi-resolution Emission Inventory in China (MEIC). Trends in Chinese emissions up to 2017 have been reported 47, and its variation during the COVID-19 pandemic in 2020 have also been described 48. Neither of those two references covered 2018, so instead the average data for 2017 and 2019 were calculated. From the MEIC estimations, the reduction in NO$_x$ emissions in
2020 compared to those in 2019 during February and March was 36% and 14%, respectively, and the reduction in SO\textsubscript{2} emissions was 27% and 11%, respectively. NH\textsubscript{3} emissions were not provided for COVID-19\textsuperscript{48}, but because their main source is agriculture, we assumed that NH\textsubscript{3} emissions were unchanged during COVID-19\textsuperscript{49} and we fixed them to the levels for 2017. Biogenic emissions were prepared from the Model of Emissions of Gases and Aerosols from Nature (MEGAN)\textsuperscript{50} using the WRF-simulated meteorological field for the corresponding period. Biomass-burning emissions were taken from the beta version of the Global Fire Emissions Database (GFED) version 4.1\textsuperscript{51}. SO\textsubscript{2} emissions from 15 volcanoes in Japan were prepared from a measurement report by the Japan Meteorological Agency (JMA)\textsuperscript{52}. Sulphur emissions from ships could influence air quality, especially in areas close to the sea\textsuperscript{53}. Because of the limited ship emission inventory, HTAP version 2.2 was used for 2018 and 2019, and a reduction in SO\textsubscript{2} emissions of 77% for 2020 and 2021 was applied based on a report by the International Maritime Organization\textsuperscript{54}. This reduction rate was consistent with a high-resolution emission inventory developed for Japan\textsuperscript{55}. To our knowledge, the emission situation in 2021—regarded as after the COVID-19 pandemic—is not yet available, so to consider the emission status in the region downwind of China from the observational facts, two experiments were conducted: (i) M21E20, which assumed the emission levels during COVID-19 for those in 2021, and (ii) M21E19, which assumed the emission levels before COVID-19 for those in 2021. By comparing these two experiments, the emission status in 2021 was investigated.

The model performance was evaluated statistically using the metrics of \( R \), \( NMB \) and \( NME \) defined as

\[
R = \frac{\sum_{i=1}^{N} \left( O_i - \bar{O} \right) \left( M_i - \bar{M} \right)}{\sqrt{\sum_{i=1}^{N} \left( O_i - \bar{O} \right)^2} \sqrt{\sum_{i=1}^{N} \left( M_i - \bar{M} \right)^2}}, \tag{3}
\]

\[
NMB = \frac{\sum_{i=1}^{N} \left( M_i - O_i \right)}{\sum_{i=1}^{N} O_i}, \tag{4}
\]

\[
NME = \frac{\sum_{i=1}^{N} | M_i - O_i |}{\sum_{i=1}^{N} O_i}, \tag{5}
\]

where \( N \) is the total number of paired observations (\( O \)) and models (\( M \)), and these averages are denoted as \( \bar{O} \) and \( \bar{M} \), respectively. The recommended metrics based on a literature review for the United States\textsuperscript{56} are given in Table 2.

**Data availability**
The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

**Declarations**

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**Author contributions**

S.I. conducted the model simulations and analysed the model results, surface observations, and satellite measurements. Y.Y. conducted the surface observations at the Tsushima site and analysed the observation dataset. Z. W. and I.U. conducted the analyses of the surface observations. S.I and I.U designed the study, and wrote the manuscript with valuable input from all co-authors. All authors reviewed the manuscript.

**Additional information**

**Competing interests**

The authors declare no competing interests.

**References**

1. Dian, D., Xing, J., Wang, S. & Hao, J. Estimated contributions of emissions controls, meteorological factors, population growth, and changes in baseline mortality to reductions in ambient PM$_{2.5}$ and PM$_{2.5}$-related mortality in China, 2013–2017. Environ. Health Perspect. **127**, 067009; 10.1289/EHP4157 (2019).

2. Zhai, S. *et al.* Fine particulate matter (PM$_{2.5}$) trends in China, 2013–2018: Separating contributions from anthropogenic emissions and meteorology. Atmos. Chem. Phys. **19**, 11031–11041 (2019).

3. Zhang, Q. *et al.* Drivers of improved PM$_{2.5}$ air quality in China from 2013 to 2017. Proc. Natl Acad. Sci. U. S. A. **116**, 24463-24469; 10.1073/pnas.1907956116 (2019).

4. Uno, I. *et al.* Paradigm shift in aerosol chemical composition over regions downwind of China. Sci. Rep. **10**, 21748; 10.1038/s41598-020-63592-6 (2020).
5. Itahashi, S., Hayami, H., Yumimoto, K. & Uno, I. Chinese province-scale source apportionments for sulfate aerosol in 2005 evaluated by the tagged tracer method. Environ. Poll. 220, 1366–1375 (2017).

6. Itahashi, S. Toward synchronous evaluation of source apportionments for atmospheric concentration and deposition of sulfate aerosol over East Asia. J. Geophys. Res. Atmos. 123, 2927–2953; 10.1002/2017JD028110 (2018).

7. Itahashi, S., Uno, I., Hayami, H. & Fujita, S. Modeling investigation of controlling factors in the increasing ratio of nitrate to non-seasalt sulfate in precipitation over Japan. Atmos. Environ. 92, 171–177 (2014).

8. Itahashi, S. et al. A 15-year record (2001-2015) of the ratio of nitrate to non-sea-salt sulfate in precipitation over East Asia. Atmos. Chem. Phys. 18, 2835–2852 (2018).

9. Huang, C. et al. Clinical features of patients infected with 2019 novel coronavirus in Wuhan, China. Lancet 395, 497–506; 10.1016/S0140-6736(20)30183-5 (2020).

10. Chinazzi, M. et al. The effect of travel restrictions on the spread of the 2019 novel coronavirus (COVID-19) outbreak. Science 368, 395–400; 10.1126/science.aba9757 (2020).

11. Tian, H. et al. An investigation of transmission control measures during the first 50 days of the COVID-19 epidemic in China. Science 368, 638–642; 10.1126/science.abb6105 (2020).

12. Liu, F. et al. Abrupt decline in tropospheric nitrogen dioxide over China after the outbreak of COVID-19. Sci. Adv. 6, eabc2992; 10.1126/sciadv.abc2992 (2020).

13. Wang, Z. et al. Impacts of COVID-19 lockdown, Spring Festival and meteorology on the NO2 variations in early 2020 over China based on in-situ observations, satellite retrievals and model simulations. Atmos. Environ. 244, 117972 (2021).

14. Wang, Y. et al. Changes in air quality related to the control of coronavirus in China: Implications for traffic and industrial emissions. Sci. Total Environ. 731, 139133 (2020).

15. Bao, R. & Zhang, A. Does lockdown reduce air pollution? Evidence from 44 cities in northern China. Sci. Total Environ. 731, 139052 (2020).

16. Chatani, S., Shimadera, H., Itahashi, S. & Yamaji, K. Comprehensive analyses of source sensitivities and apportionments of PM2.5 and ozone over Japan via multiple numerical techniques. Atmos. Chem. Phys., 20, 10311–10329 (2020).

17. Liu, J., Li, J. & Yao, F. Source-receptor relationship of transboundary particulate matter pollution between China, South Korea and Japan: Approaches, current understanding and limitations. Crit. Rev. Environ. Sci. Technol.; 10.1080/10643389.2021.1964308ERL (2021).

18. International Monetary Fund. World economic outlook: Recovery during a pandemic—Health concerns, supply disruptions, price pressures. Report at https://www.imf.org/-/media/Files/Publications/WEO/2021/October/English/text.ashx (2021).

19. Chatani, S., Itahashi, S. & Yamaji, K. Advantages of continuous monitoring of hourly PM2.5 component concentrations in Japan for model validation and source sensitivity analyses. Asian. J.
20. Liu, T., Chan, A. W. H. & Abbatt, J. P. D. Multiphase oxidation of sulfur dioxide in aerosol particles: implications for sulfate formation in polluted environments. Env. Sci. Tech. 55, 4227–4242 (2021).

21. Uno, I. et al. Seasonal variation of fine- and coarse-mode nitrates and related aerosols over East Asia: Synergetic observations and chemical transport model analysis. Atmos. Chem. Phys. 17, 14181–14197 (2017).

22. Itahashi, S. et al. Model performance differences in fine-mode nitrate aerosol during wintertime over Japan in the J-STREAM model inter-comparison study. Atmosphere 11, 511 (2020).

23. Itahashi, S. et al. Model estimation of sulfate aerosol sources collected at Cape Hedo during an intensive campaign in October-November, 2015. Aerosol Air Qual. Res. 17, 3079–3090 (2017).

24. Yamamura, Y. et al. Contribution of volcanic SO$_2$ emission to high concentration PM$_{2.5}$ under summertime Pacific High condition. J. Jpn. Soc. Atmos. Environ. 55, 169–180 (2020) (in Japanese with English abstract).

25. Bleeker, A., Hicks, W. K., Dentener, F., Galloway J. & Erisman, J. W. N deposition as a threat to the World’s protected area under the Convention of Biological Diversity. Env. Poll. 159, 2280–2288 (2011).

26. Sutton, M.A. & Bleeker, A. The shape of nitrogen to come. Nature 494, 435–437 (2013).

27. Itahashi, S., Hayami, H., Uno, I., Pan, X. & Uematsu, M. Importance of coarse-mode nitrate produced via sea salt as atmospheric input to East Asian oceans. Geophys. Res. Lett. 43, 5483–5491 (2016).

28. Itahashi, S. et al. MICS-Asia III: overview of model intercomparison and evaluation of acid deposition over Asia. Atmos. Chem. Phys. 20, 2667–2693; 10.5194/acp-20-2667-2020 (2020).

29. Itahashi, S. et al. Nitrogen burden from atmospheric deposition in East Asian oceans in 2010 based on high-resolution regional numerical modeling. Env. Poll. 486, 117309 (2021).

30. Ge, B. et al. Model Inter-Comparison Study for Asia (MICS-Asia) phase III: Multimodel comparison of reactive nitrogen deposition over China. Atmos. Chem. Phys. 20, 10587–10610; 10.5194/acp-20-10587-2020 (2020).

31. Itahashi, S. et al. Inverse estimation of NO$_x$ emissions over China and India 2005–2016: contrasting recent trends and future perspectives. Env. Res. Lett. 14, 124020 (2019).

32. Nickolay A. et al. OMI/Aura NO$_2$ Cloud-Screened Total and Tropospheric Column L3 Global Gridded 0.25 degree × 0.25 degree V3. NASA Goddard Space Flight Center, Goddard Earth Sciences Data and Information Services Center (GES DISC) https://doi.org/10.5067/Aura/OMI/DATA3007 (2019).

33. Ministry of the Environment. http://www.env.go.jp/air/%20osen/pm_resultmonitoring/post_25.html (n.d.) (in Japanese).

34. Itahashi, S. et al. Nitrate transboundary heavy pollution over East Asia in winter. Atmos. Chem. Phys. 17, 3823–3843 (2017).

35. Lai, S. et al. Effect of non-pharmaceutical interventions to contain COVID-19 in China. Nature 585, 410–413; 10.1038/s41586-020-2293-x (2020).
36. Cabinet Secretariat of Japan. COVID-19 information and resources. https://corona.go.jp/en/ (n.d.)
37. One World Data. COVID-19: Stringency index. https://ourworldindata.org/covid-stringency-index (2020).
38. Ramboll Environment and Health. *User's guide, comprehensive air quality model with extensions version 6.40* (Ramboll, 2016).
39. Skamarock, W. C. *et al.* *A description of the advanced research WRF version 4.* NCAR Tech. Note, NCAR/TN-556+STR. 162 (National Center for Atmospheric Research, Boulder, 2019)
40. Carter, W.P.L. Development of the SAPRC-07 chemical mechanism. Atmos. Environ., 44, 5324–5335 (2010).
41. Hutzell, W. T., Luecken, D. J., Appel, W. K. & Carter, W. P. Interpreting predictions from the SAPRC07 mechanism based on regional and continental simulations. Atmos. Environ. 46, 417–429 (2012).
42. Itahashi, S. *et al.* Model performance differences in sulfate aerosol in winter over Japan based on regional chemical transport models of CMAQ and CAMx. Atmosphere 9, 488 (2018).
43. Itahashi, S., Uchida, R., Yamaji, K. & Chatani, S. Year-round modeling of sulfate aerosol over Asia through updates of aqueous-phase oxidation and gas-phase reactions with stabilized Criegee intermediates. Atmos. Environ. X. 12, 100123 (2021).
44. Zheng, B. *et al.* Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China. Atmos. Chem. Phys. 15, 2031-2049; 10.5194/acp-15-2031-2015 (2015).
45. US EPA. Hemispheric CMAQ model version 5.3beta output data – 2016 seasonally averaged 108km for N. hemisphere, UNC Dataverse, V1. Dataset at https://doi.org/10.15139/S3/QJDYWO (2019)
46. Janssens-Maenhout, G. *et al.* HTAP_v2.2: A mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution. Atmos. Chem. Phys. 15, 11411–11432 (2015).
47. Zheng, B. *et al.* Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions. Atmos. Chem. Phys. 18, 14095–14111; 10.5194/acp-18-14095-2018 (2018).
48. Zheng, B. *et al.* Changes in China's anthropogenic emissions and air quality during the COVID-19 pandemic in 2020. Earth Syst. Sci. Data 13, 2895–2907; 10.5194/essd-13-2895-2021 (2021).
49. Forster, P. M. *et al.* Current and future global climate impacts resulting from COVID-19. Nat. Clim. Change 10, 913–919 (2020).
50. Guenther, A. B. *et al.* The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1). An extended and updated framework for modeling biogenic emissions. Geosci. Model Dev. 5, 1471–1492 (2012).
51. van der Werf, G. R. *et al.* Global fire emissions estimates during 1997–2016. Earth Syst. Sci. Data 9, 697–720 (2017).
52. Japan Meteorological Agency. http://www.data.jma.go.jp/svd/vois/data/tokyo/volcano.html (n.d.) (in Japanese).
53. Itahashi, S., Sakurai, T., Shimadera, H., Araki, S. & Hayami, H. Long-term trends of satellite-based fine-mode aerosol optical depth over the Seto Inland Sea, Japan, over two decades (2001-2020). Env. Res. Lett. 16, 064062; 10.1088/1748-9326/ac03db (2021).

54. International Maritime Organization. Air pollution and energy efficiency: Study on the effects of the entry into force of the global 0.5% fuel oil sulphur content limit on human health. https://edocs.imo.org/Final Documents/English/MEPC 70-INF.34 (E).docx (2016).

55. Sakurai, T., Ito, M. & Hanayama, S. Development of air pollutants emission inventories for ships around Japan on a high geographical resolution. Asian J. Atmos. Environ. 15, 2020096; 10.5572/ajae.2020.096 (2021).

56. Emery, C. et al. Recommendations on statistics and benchmarks to assess photochemical model performance. J. Air Waste Manag. Assoc. 67, 582–598; 10.1080/10962247.2016.1265027 (2017)

**Figures**

![Satellite-measured tropospheric NO₂ column density over East Asia](image_url)

**Figure 1**

Satellite-measured tropospheric NO₂ column density over East Asia: (a) before COVID-19; (b) difference between during COVID-19 and before COVID-19; (c) difference between after COVID-19 and during COVID-19. The inset in the upper left of (b) and (c) shows the absolute values during and after COVID-19, respectively. The inset in the lower right of (c) shows the three observation sites analysed in this study.
Figure 2

Two-month (February and March) average of (left) PM$_{2.5}$, (centre) SO$_4^{2-}$ and (right) NO$_3^-$ concentrations from 2018 to 2021 at (top) Goto, (middle) Tsushima and (bottom) Oki. For each year, the observation is shown on the left and model is shown on the right. For 2021, the model experiments of M21E20 and M21E19 are shown by the lighter colour and darker colour and denoted as E20 and E19, respectively.
Figure 3

Changes in (left) PM$_{2.5}$, (centre) SO$_4^{2-}$ and (right) NO$_3^-$ concentrations during and after COVID-19 at (top) Goto, (middle) Tsushima and (bottom) Oki. These were calculated from the mean values in Fig. 2. For each year, the observation is shown on the left and the model is shown on the right. For 2021, the model experiments of M21E20 and M21E19 are shown by the lighter colour and darker colour and denoted as E20 and E19, respectively.

Figure 4
Changes in (a) PM$_{2.5}$, (b) SO$_4^{2-}$ and (c) NO$_3^-$ concentrations during and after COVID-19 due to meteorology and changes in natural emissions by calculating the M21E20 values minus the M20E20 ones. Modeled 10-m wind field (color denotes 10-m wind speed) in (d) 2020 (M20) and (e) 2021 (M21), and change in 10-m wind field by calculating the M21 WRF meteorological field minus the M20 one.

Figure 5
(Left) Observed and (right) modeled scatterplots of $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ at (top) Goto, (middle) Tsushima and (bottom) Oki. The number near each circle indicates the year (i.e. ‘18’ means 2018). At Goto, the results from a previous study$^4$ are shown in grey. The previous study analysed PM$_1$ components, whereas the present study analysed PM$_{2.5}$ components. For the model results, those of experiment M21E20 are shown by the grey closed circles.

**Supplementary Files**

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