Paradigm shift in aerosol chemical composition over regions downwind of China

Itsushi Uno1*, Zhe Wang1,2*, Syuichi Itahashi3, Keiya Yumimoto1, Yuki Yamamura4, Ayako Yoshino5, Akinori Takami5, Masamitsu Hayasaki6 & Byung-Gon Kim7

A rapid decrease in PM$_{2.5}$ concentrations in China has been observed in response to the enactment of strong emission control policies. From 2012 to 2017, total emissions of SO$_2$ and NO$_x$ from China decreased by approximately 63% and 24%, respectively. Simultaneously, decreases in the PM$_{2.5}$ concentration in Japan have been observed since 2014, and the proportion of stations that satisfy the PM$_{2.5}$ environmental standard (daily, 35 µg/m$^3$; annual average, 15 µg/m$^3$) increased from 37.8% in fiscal year (FY) 2014 (April 2014 to March 2015) to 89.9% in FY 2017. However, the quantitative relationship between the PM$_{2.5}$ improvement in China and the PM$_{2.5}$ concentration in downwind regions is not well understood. Here, we (1) quantitatively evaluate the impacts of Chinese environmental improvements on downwind areas using source/receptor analysis with a chemical transport model, and (2) show that these rapid emissions reductions improved PM$_{2.5}$ concentrations both in China and its downwind regions, but the difference between SO$_2$ and NO$_x$ reduction rates led to greater production of nitrates (e.g., NH$_4$NO$_3$) due to a chemical imbalance in the ammonia–nitric acid–sulfuric acid–water system. Observations from a clean remote island in western Japan and numerical modeling confirmed this paradigm shift.

The long-range trans-boundary transport behavior of pollutants in East Asia is an important environmental issue due to frequent outflows of heavy pollution. Among pollutants, PM$_{2.5}$ (particulate matter less than 2.5 µm in diameter) poses serious human health risks, including lung cancer, respiratory disease, and asthma, particularly over China and its downwind regions1-3. Serious PM$_{2.5}$ pollution has been observed in the northern China region since the early 2010s. To reduce this pollution, China has implemented active clean air policies in recent years (e.g., the Action Plan for Prevention and Control of Air Pollution, enacted in September 2013). These plans include the phasing out of outdated industrial capacity, small high-emission factories, and small coal-fired industrial boilers, as well as the strengthening of emission standards for power plants, industries, and vehicles, and the replacement of residential coal use with electricity and natural gas4-6. These strong emission-reduction policies in China have led to a successful reduction in PM$_{2.5}$ concentration (e.g., PM$_{2.5}$ concentrations decreased from 102 µg/m$^3$ in 2013 to 43 µg/m$^3$ in 2019, as observed at the U.S. embassy in Beijing). Rapid reductions in SO$_2$ and NO$_x$ emissions were also confirmed using environmental satellite data8 and bottom-up emissions inventory studies1. MODIS AOD (aerosol optical depth) data revealed that there has been a consistent trend of year-to-year decreases in China and its downwind regions. The chemical composition of PM$_{2.5}$ also changed significantly over China, with especially large decreases in levels of organic matter, mineral components, and sulfate aerosols9. Studies have suggested that emission control has a dominant effect on PM$_{2.5}$ reduction compared to inter-annual meteorological variation10-12. Further research using bottom-up emission inventory and numerical models revealed that different emission control measures contribute to reductions in PM$_{2.5}$6,13. Meanwhile, recent studies have reported significant health benefits resulting from PM$_{2.5}$ improvements in China14,15.
Focusing on regions downwind of China, the proportion of monitoring stations meeting the Japanese environmental standard for PM$_{2.5}$ (defined as the achievement ratio) increased rapidly from 37.8% in fiscal year (FY) 2014 to 89.9% in FY 2017 (Ministry of Environment, Japan)\textsuperscript{16}. During this period, Japanese pollutant emissions exhibited a slight decreasing trend \textsuperscript{17}. Improvements in the PM$_{2.5}$ achievement ratio result from complex interactions between Japanese and Chinese emission-reduction measures, and it is not yet clear which factor has a greater contribution. This lack of clarity arises because most PM$_{2.5}$ studies are focused on an individual country, and quantitative evaluations or correlation analyses of the impacts of environmental improvement on downwind regions (beyond national borders) have rarely been published\textsuperscript{18–21}. Studying the relationship between decreased PM$_{2.5}$ concentrations in China and its downwind regions and clarifying quantitative source/receptor (S/R) relationships are essential for establishing better environmental policies.

Remote island observations of aerosol chemical compositions off the west coast of Japan (eastern edge of the East China Sea) indicate that sulfate concentrations have decreased significantly, consistent with the decrease in SO$_2$ emissions in China. Although NO$_x$ emissions also decreased, the observed nitrate concentration increased continuously in recent years\textsuperscript{22}. This increase in the nitrate concentration could lead to excess N input to the oceans surrounding East Asia, which may have impacts on the marine ecosystem\textsuperscript{23}. In this paper, we support this observational finding with chemical transport model (CTM) sensitivity experiments, in which SO$_2$ and NO$_x$ emissions are reduced at different rates to confirm the observed trend. These experiments showed that the observed nitrate increase can be explained by changes in the ammonia–nitric acid–sulfuric acid–water system balance due to the greater rate of decrease in SO$_2$ emissions compared to that in NO$_x$ emissions.

Results
We analyzed hourly surface-level PM$_{2.5}$ observational data from Japan and China and calculated the annual average PM$_{2.5}$ concentration for each region. For Korea, we used annual average values from the Air Korea website. The observational data used in this study are described in the Methods section.

Satellite observations of NO$_x$ and SO$_2$ from the Ozone Monitoring Instrument (OMI) were also used for the analysis of emission trends between 2005 and 2019. Gridded (0.25 $\times$ 0.25 degree) Level 3 data from NASA were used for this study and annually-averaged data were used to examine year-to-year trends over China, Korea, and Japan (NASA OMI website)\textsuperscript{24}.

Figure 1 shows the annual average PM$_{2.5}$ concentrations over Fukuoka, Japan and Beijing (at the U.S. Embassy, China (Supplementary Figure S1 shows the locations of the observation sites). This figure includes estimated SO$_2$, NO$_x$, and NH$_3$ emissions over China\textsuperscript{1}, and tropospheric vertical column densities (VCDs) of SO$_2$ and NO$_2$ over central eastern China (CEC) (OMI satellite data from 2011 to 2019)\textsuperscript{24}. This figure also shows the PM$_{2.5}$ achievement ratio for Japan. Similar plots for the average of 74 cities in China and Korea (including two background sites), and several remote Japanese sites are provided in Supplementary Figure S2. Supplementary Figure S3 shows the annual average VCDs of SO$_2$ and NO$_2$ levels in East Asian regions between 2011 and 2019 based on satellite retrieval data. Supplementary Figure S4 shows the year-to-year average trends in three regions (CEC, Korea, and Japan) for SO$_2$ and NO$_2$ levels based on satellite observations.

The PM$_{2.5}$ trends in Beijing and the average trend of 74 Chinese cities were very strongly correlated (Supplementary Figure S2(a)), although the PM$_{2.5}$ concentration in Beijing was 40% higher than the average concentration of 74 cities in 2013 (this difference became negligible in 2018, as the rate of decrease in Beijing was greater). This trend was quite consistent with that in Japan (Supplementary Figure S2(c)). The correlation coefficient (R) between Beijing and Fukuoka was 0.98. The average PM$_{2.5}$ trend for Korea (Supplementary Figure S2(b)) also showed a generally decreasing trend but with a slight difference from those in China and Japan up to 2017.

Bottom-up inventory results and satellite data also exhibit good correlations (Fig. 1). OMI SO$_2$ observations (Supplementary Figure S3) indicated that a rapid decrease in SO$_2$ was achieved over the CEC area, and the color
representing SO₂ in the image has been nearly absent since 2017 (using the same color scale). We found that the \( \text{SO}_2 \) VCDs in Korea and Japan exhibited small decreases or constant levels after 2010 (see Supplementary Figure S4), whereas \( \text{NO}_2 \) VCDs in Korea and Japan remained almost constant and increased slightly, respectively, since 2017. \( \text{NO}_2 \) VCDs exhibited an increasing trend over CEC after 2016.

Figure 2 shows the horizontal distribution of annually-averaged PM\(_{2.5}\) concentrations over Japan between 2013 and 2018. Numbers in parentheses indicate PM\(_{2.5}\) achievement ratios for Japan.

![Horizontal distribution of annually averaged PM\(_{2.5}\) concentrations over Japan between 2013 and 2018](image)

Figure 3 is similar to Fig. 2 but covers an extended region including Eastern China and Korea. Different colors in the figure represent different rates of decrease in observed PM\(_{2.5}\) levels, defined as:

\[
\Delta \text{PM}_{2.5} = \frac{(\text{PM}_{2.5-2015} - \text{PM}_{2.5-\text{yyyy}})}{\text{PM}_{2.5-2015}},
\]

(1)
where yyyy indicates a year. The rate was scaled based on the concentration in 2015. Negative values of $\Delta PM_{yyyy}$ indicate that PM$_{2.5}$ increased relative to 2015, as seen at some Korean sites. The annual average $\Delta PM_{yyyy}$ values over Eastern China (regionally averaged over 115°–123° E, 28°–43° N) from 2016 to 2018 were 0.120 ± 0.054, 0.191 ± 0.077, and 0.262 ± 0.094, respectively. This result indicates that the observed average PM$_{2.5}$ concentration over Eastern China decreased by approximately 7% annually between 2016 and 2018.

The detailed changes over Japan were discussed in the context of Fig. 2. Figure 3 shows observed annual average rates of decrease (regionally averaged over 130°–142° E, 33°–37° N) of 0.104 ± 0.046, 0.151 ± 0.062, and 0.172 ± 0.064 since 2015. $\Delta PM_{yyyy}$ values for both the Korean average and individual stations are shown. A complicated variation in $\Delta PM_{yyyy}$ was observed, with some stations exhibiting positive changes or different variation patterns between years, except at the upwind background stations in Baengyeongdo and Jeju, which will be discussed later.

We used the 3-D Goddard Earth Observing System chemical transport model (GEOS-Chem) for emission sensitivity analysis, including that of the S/R relationship for PM$_{2.5}$. Details of the GEOS-Chem settings and S/R analysis are described in the Methods section.

The model results were analyzed to obtain S/R values. We confirmed that the annual average contribution of Japanese domestic emissions to Fukuoka PM$_{2.5}$ was approximately 20%, and the Chinese contribution was approximately 60% based on the meteorological conditions in 2014. S/R results are very useful for evaluating possible strategies for improving PM$_{2.5}$ levels over downwind regions after enacting appropriate emission controls.
in one region. For example, the contribution of PM$_{2.5}$ from China (mainly from northern China) to Fukuoka was approximately 60%, and thus if the PM$_{2.5}$ concentration in China decreases by 40% (e.g., from 100 to 60 μg/m$^3$ between 2014 and 2017), the decrease in PM$_{2.5}$ concentration in Fukuoka can be calculated as follows: 60% × 40% = 24% (assuming that all emissions except those from China remain constant). The observed decrease in PM$_{2.5}$ concentration in Fukuoka (18.5 to 14.5 μg/m$^3$) was 22%, which is in good agreement with the model-based S/R estimate.

The contour lines in Fig. 3 represent S/R responses and were calculated by multiplying the fraction of the PM$_{2.5}$ contribution from China at each point by the rate of decrease in Chinese PM$_{2.5}$ concentration. The contours in Fig. 3(a–c) represent rates of decrease over East Asia in response to different decreases in PM$_{2.5}$ concentration in China. The rate of decrease in PM$_{2.5}$ concentration in China was set to 12%, 19%, or 26% based on observations. The observed relationship between rates of decrease in China and Japan can be explained using these contour lines.

For Korea, trends at the upwind background sites of Baengyeongdo and Jeju showed a consistent decreasing signal, in agreement with the decrease based on the S/R relationship. However, trends at other Korean sites cannot be explained by the S/R contour lines, and some cities (e.g., Seoul) exhibited significant increases in PM$_{2.5}$ concentrations in 2016 and 2017 compared with 2015 and large year-to-year variations.

OMI SO$_2$ and NO$_2$ variations across Korea cannot explain the observed changes in the PM$_{2.5}$ concentration. In Korea, the SO$_2$ VCD exhibited a small decrease or no change after 2010, and the NO$_2$ VCD remained almost unchanged (see Supplementary Figure S4); thus, the trend of the average PM$_{2.5}$ was not clearly correlated with the local emissions pattern (particularly in urban areas). Elucidating these patterns and their drivers in Korea is a subject for future research.

We examined recent PM$_{2.5}$ and aerosol composition changes over a clean and remote island, Fukue Island, which is located at the western edge of the Japanese mainland and eastern edge of the East China Sea (see Supplementary Figure S1). Details of the observations from Fukue Island and comparison with the GEOS-Chem simulation can be found in the Methods section.

Figure 4 shows observation results from Fukue Island averaged between February and April. Figure 4(a) shows the aerosol composition ratios among chloride, NO$_3^-$, SO$_4^{2-}$, NH$_4^+$, and organic aerosols. Figure 4(b) shows a scatter diagram of SO$_4^{2-}$ and NO$_3^-$ for each year.

As shown in Fig. 4(a), the observed sulfate concentration decreased significantly (by 40%) at Fukue Island. This result is consistent with the decrease in SO$_2$ emissions over China. Although NO$_x$ and NH$_3$ emissions were also reduced, the observed nitrate concentrations increased continuously. This result could be explained by the chemical balance of the ammonia–nitric acid–sulfuric acid–water system. This thermodynamic equilibrium process is included in the GEOS-Chem simulation described in the Methods section, which allows for detailed studies on chemical balance. Due to the extremely low vapor pressure of sulfuric acid, sulfuric acid produced in the atmosphere consumes ammonia and is neutralized, forming ammonium sulfate aerosol. Then, the leftover ammonia, referred to as free ammonia, is available for the potential formation of ammonium nitrate. As a result, the reduction of sulfuric acid causes more free ammonia to be available, leading to the formation of more ammonium nitrate. Seinfeld and Pandis (2016) indicates that about half of the decrease in concentration of (NH$_3$)$_2$SO$_4$ will be offset by the increase in NH$_4$NO$_3$. The relationship between the decrease in sulfate and increase in nitrate depends primarily on the concentrations of their precursors, relative humidity (RH), and temperature.

Although SO$_2$, NO$_x$, and NH$_3$ emissions over China have all been reduced, the decrease in NO$_x$ is significantly smaller than that in SO$_2$, and the decrease in NH$_3$ is much smaller than that in either SO$_2$ or NO$_x$ (see Fig. 1). If the increase in nitrate due to SO$_2$ reduction is larger than the nitrate decreases due to decreases in NO$_x$ and NH$_3$ emissions, the overall effects of emission control will lead to increased nitrate concentration. These phenomena were actually observed on Fukue Island from 2012 to 2019 (as seen in Fig. 4(b)), where sulfate decreased by 1.7 μg/m$^3$, while nitrate increased by 1.7 μg/m$^3$, causing the NO$_3^-$ concentration to increase by almost four-fold compared to the 2012–2014 period. A more detailed analysis of these phenomena based on the GEOS-Chem model is described below.

**Discussion**

To quantitatively analyze the increase in NO$_3^-$, we modeled an additional four cases of sensitivity experiments, changing the SO$_2$ and NO$_x$ emission intensities based on the bottom-up Multi-resolution Emission Inventory for China (MEIC) results (Table 1). Emission reduction was applied only in the China region, and emissions in all other regions were the same as in the control experiment.

CNTL (S10N10) was the control experiment. Case S04N08 was designed based on the MEIC emission reduction rate and OMI satellite changes, and thus is suitable for examining recent emission changes. Case S07N09 was designed to examine the linearity of decreases in SO$_2$ (between the S10, S07, and S04 cases), and case S04N10 was designed to examine NO$_x$ sensitivity under a constant SO$_2$ condition (with S04N08). In this sensitivity study, emissions of NH$_3$ and non-methane volatile organic compounds were the same as in CNTL.

The results of the modeled sensitivity experiments are shown in Fig. 4(c) for SO$_4^{2-}$ and NO$_3^-$ in CEC, the centers of the Yellow Sea and East China Sea, and Fukue Island (model results were averaged between February and April for consistency with observations). Note that Fig. 4(c) demonstrates the typical response of the ammonia–nitric acid–sulfuric acid–water system to the emission sensitivity shown in Table 1. Thus, the absolute concentration level is different from the ACSM observation, but the fundamental changes observed can be explained by the model emission sensitivity experiment.

As shown in Fig. 4(c), the sensitivity experiment between CNTL (=S10N10) and S04N10 for CEC showed decreased SO$_4^{2-}$ (ΔSO$_4^{2-}$ = −4.5 μg/m$^3$) and increased NO$_3^-$ (ΔNO$_3^-$ = +2.2 μg/m$^3$), consistent with the response discussed in the Results section. The series of sensitivity experiments (CNTL, S07N09 and S04N08) shows a nearly linear response between ΔSO$_4^{2-}$ and ΔNO$_3^-$ even with decreasing NO$_x$ emission. The differing
Figure 4. (a) Year-to-year changes in aerosol composition observed at Fukue Island (averaged from February to April), (b) scatter plot of averaged SO$_4^{2-}$ and NO$_3^{-}$ concentrations at Fukue Island from observation data and (c) GEOS Chem sensitivity analysis (extracted for CEC, Yellow Sea, East China Sea, and Fukue Island). Model results were averaged from February to April.

| Case Code   | Purpose                      | SO$_2$ emission | NO$_x$ emission |
|-------------|-------------------------------|-----------------|-----------------|
| CNTL (S10N10) | Control experiment           | 100%            | 100%            |
| S04N08      | Decrease relative to Fig. 1   | 40%             | 80%             |
| S07N09      | SO$_2$ linearity              | 70%             | 90%             |
| S04N10      | NO$_x$ sensitivity            | 40%             | 100%            |

Table 1. Design of a model for sensitivity analysis of Chinese emissions.
responses of NO$_3^-$ in S04N08 and S04N10 (NO$_x$ emission difference) is most apparent for Beijing. This difference becomes very small over the downwind regions of the East China Sea and Fukue Island. The ratio of $\Delta$SO$_4^{2-}$: $\Delta$NO$_3^-$ ranges from 1:0.5 to 1:0.65, becoming larger as the transport distance increases. As noted above, the relationship between the decrease of sulfate and the increase of nitrate is strongly dependent on RH, temperature and a heterogeneous reaction with sea salt (for NaNO$_3$ formation) during transport from mainland China over the ocean. These responses are quite consistent with observations at Fukue Island.

The change in NH$_3$ concentration between CNTL and S04N08 is of great interest, and this result is shown in Supplementary Figure S5. The NH$_3$ concentration in the S04N08 experiment was more than double that over CEC (i.e., increase in free NH$_3$) and NH$_3$ concentration increases were simulated over western Japan, including Fukue Island. The changes in NH$_3$ concentration over CEC were also supported by Infrared Atmospheric Sounding Interferometer (IASI) satellite observations. The conclusions of these sensitivity studies were reasonable, showing that reductions in SO$_2$ emissions change the balance of the ammonia–nitric acid–sulfuric acid–water system, creating free NH$_3$ that reacts with HNO$_3$ to form NH$_4$NO$_3$, which is transported to downwind regions, especially in the cold season.

Figure 5 shows the horizontal distribution of scaled annual mean $\Delta$SO$_4^{2-}$ and $\Delta$NO$_3^-$ from the model sensitivity study, based on the CNTL and S04N08 experiments. These indices are calculated as follows:

\[
\Delta SO_4^{2-} = \frac{(SO_4^{2-}_{S04N08} - SO_4^{2-}_{CNTL})}{SO_4^{2-}_{CNTL}} \quad (2a)
\]

\[
\Delta NO_3^- = \frac{(NO_3^-_{S04N08} - NO_3^-_{CNTL})}{NO_3^-_{CNTL}} \quad (2b)
\]

The SO$_4^{2-}$ decrease ($\Delta$SO$_4^{2-}$) over mainland China exceeded −50%, consistent with the 60% decrease in SO$_2$ (Fig. 1a). Over western and eastern Japan, SO$_4^{2-}$ decreased by 30% and 20%, respectively. We found that this rate of decrease was linearly proportional to the SO$_2$ reduction rate within China via a comparison with case S07N09. The impacts of the decrease in SO$_2$ in China clearly covered a large area downwind.

For NO$_3^-$, $\Delta$NO$_3^-$ over China was not significant, which is consistent with recent observations. Several areas downwind of China exhibited increased NO$_3^-$. Over the East China Sea, the rate of increase in NO$_3^-$ exceeded 90%, as NO$_3^-$ concentrations were low in this area in the CNTL experiment; thus, a small increase in NO$_3^-$ results in a large rate of increase. In the Fukue Island region, this increase was approximately 60%. Figure 5 shows annually averaged values, and ratios increased when averaged over the cold season (February to April) because NH$_4$NO$_3$ is more stable in cold weather, as discussed below.

Figure 6 shows time–longitude trends of the NO$_3^-$ increase from experiments CNTL to S04N08 along the latitude of 32.5° N. This latitude corresponds the typical transport route from Shanghai to Fukue Island. The increase in concentrations over the downwind regions of China between December and March was significant and was caused by cold weather increasing NH$_4$NO$_3$ stability. During the warm season, the transport path changes and warm temperatures cause NH$_4$NO$_3$ aerosols to enter the gas phase as HNO$_3$. At Fukue Island, the increase reached 1 µg/m$^3$ in winter, consistent in magnitude with observations at Fukue Island. Notably, the eastern edge of area in which NO$_3^-$ increased (on the order of 0.5 µg/m$^3$) approaches 134° E to 136° E, where large cities such as Osaka are located. Note that the changes in concentration were small and usually difficult to detect from observations.
over urban areas of mainland Japan due to large local NOx emissions. However, this small increase may contribute significantly to the presence of excess nitrogen over the downwind region in East Asia23.

Recent studies have described how a decrease in PM2.5 can enhance the lifetime of OH radicals and increase the O3 level30 (followed by increases in the atmospheric oxidation capacity and NO3− formation). This is a reasonable mechanism that might increase the NO3− formation. However, our version of the GEOS-Chem model does not include the heterogeneous interaction between PM2.5 and OH. Our results explain the observed SO42−/NO3− changes exactly, and this indicates that a change in the atmospheric oxidation capacity is not the primary reason for the observed changes in SO42−/NO3−.

We analyzed the PM2.5 observation data from 2014 to 2019 over Japan, Korea, and China, and found that there was a clear decreasing trend over Japan, which was strongly correlated with levels in China. An emission sensitivity study based on the GEOS-Chem chemical transport model was carried out to quantify the relationship between emission levels in China and PM2.5 concentrations over downwind regions. The model results showed that the trend of an annual decrease in PM2.5 in Japan was explained primarily by reduced PM2.5 concentrations in China. We also used this model to quantitatively evaluate the impact of Chinese environmental improvements on downwind areas using S/R analysis. Rapid emission reductions played an important role in reducing PM2.5 concentrations, but a chemical imbalance in the ammonia–nitric acid–sulfuric acid–water system caused an increase in long-range NO3− transport to downwind regions. Observations on a clean remote island and numerical modeling confirmed that this paradigm shift has occurred since 2014–2015. Concentrations of sulfate, a chemical that undergoes long-range transport, are decreasing, whereas those of nitrate, which is subject only to short-distance transport, are increasing. This increase in nitrate could lead to an excess nitrogen burden in East Asia and the surrounding oceanic regions31,32. We found that the most recent satellite NO2 and SO2 VCDs, for 2019 (see Supplementary Figure S4), revealed that this paradigm shift is accelerating (because SO2 is still decreasing, whereas NO2 is now increasing), indicating that there is a need for careful continuous observation of changes in aerosol chemical compositions, both in China and the downwind regions of Japan and Korea.

Methods

Surface PM2.5 observation data. For Japan, hourly PM2.5 observation data from the Atmospheric Environmental Regional Observation System33 (AEROS; also referred to as ‘Soramame-kun’) were used for calculating annually averaged PM2.5 concentrations from 2013 to 2018. A total of 662 AEROS sites were used in this study, and after quality control processing, the AEROS data were interpolated into a 0.375° longitude-latitude grid. Data averaged across Japan were obtained from the Ministry of Environment of Japan.

For China, PM2.5 concentration data were obtained from the China National Environmental Monitoring Center34. In China, PM2.5 concentrations have been monitored in 74 major cities since the end of 2012, including cities in the Beijing-Tianjin-Hebei region, Yangtze River Delta, and Pearl River Delta, as well as Chongqing municipalities and all provincial capitals. Data from these 74 cities were collected between 2013 and 2018, averaged, and used for analysis in this study. We also analyzed PM2.5 observations taken at the U.S. Embassy in Beijing from 2011 to 2019.

For Korea, the annual average PM2.5 values were obtained from the official Air Korea website of the Ministry of Environment of Korea35. We selected nine sites, including the cities of Seoul, Busan, Gwangju, Gwangneung, Daegu, Daejeon and Mokpo, as well as Jeju and the background site of Baengnyeongdo (the locations of the latter two sites are shown in Supplementary Figure S1) for analysis between 2015 and 2018. Data collected from multiple points within large cities were averaged.

We used observation sites with more than 250 days of qualified observations.

Chemical transport model and S/R analysis. We used the GEOS-Chem model for analysis25. The model was run using the full GEOS-Chem NOx-Ox-VOC-HOx-CO chemistry option to simulate the formation of aerosols, including mineral dust, sea salt, black carbon (BC), organic carbon (OC), and secondary inorganic aerosols. The GEOS-Chem model used ISORROPIA-II36 to calculate the detailed thermodynamic equilibrium...
processes for the H\(^+\)--NH\(_4\)^+--K\(^+\)--Ca\(^{2+}\)--Mg\(^{2+}\)--Na\(^+\)--OH --SO\(_4^{2-}\)--NO\(_3^-\)--Cl --H\(_2\)O aerosol system. The model used the assimilated meteorological fields from GEOS of the NASA Global Modeling and Assimilation Office. The model has a horizontal resolution of 2\(^\circ\) × 2.5\(^\circ\) for global runs, and 0.5\(^\circ\) × 0.667\(^\circ\) for Asian one-way nesting runs (11\(^\circ\)S–55\(^\circ\)N, 70–150\(^\circ\)E), both containing 47 vertical levels from the surface to 0.01 hPa. We used anthropogenic emissions data from the Emission Database for Global Atmospheric Research\(^{37}\) for the global domain and from the Regional Emission Inventory in Asia (REAS; ver. 2.1) for the Asian domain\(^{38}\). REAS NH\(_3\) emissions data were modified to include seasonal variations in China\(^{39}\). PM\(_{2.5}\) concentrations from the model were calculated by summing the concentrations of relevant aerosols (BC, OC, SO\(_4^{2-}\), NO\(_3^-\), NH\(_4^+\), dust, and sea salt). Model simulation was conducted from the beginning of December 2013 to the end of July 2019, and the results from the first 8 months were used for model training. We primarily used the S/R model results for 2014 (when the pollution level was high) and assumed that the model results would be similar for the meteorology of different years. Other basic numerical settings were as reported in Uno et al.\(^{40,41}\). We set 19 source regions (including Japan, Korea, northern China, and central China) for S/R analysis\(^{46}\). We used a 20% reduction of emission (not zero emission) to avoid undesirable non-linearity of the chemical reactions. The source contribution from region A is calculated as follows:

\[
\text{Source contribution from region A} = \frac{(\text{PM}_{100,\text{all}} - \text{PM}_{80,\text{A}})}{\text{PM}_{100,\text{all}} - \text{PM}_{80,\text{all}}},
\]

where PM\(_{100,\text{all}}\) is the PM\(_{2.5}\) concentration under the 100% emission scenario for all regions.

Aerosol observations using the ACSM at Fukue Island. The chemical compositions and mass concentrations of atmospheric fine aerosols, i.e., fine particulate matter (PM\(_{1}\)), were observed at the remote island of Fukue, Nagasaki Prefecture, Japan (32.75° N, 128.68° E; see Supplementary Figure S1). The population on this island is approximately 40,000 and it is generally considered to have few emission sources. Aerosol chemical composition was measured using a quadrupole-type ACSM (Q-ACSM; Aerodyne Research Inc., Billerica, MA, USA). The mass concentrations of PM\(_{2.5}\) were obtained from an air-pollution monitoring station at Goto (located this island), which is the site of municipal government offices for Nagasaki prefecture. The chemical compositions and mass concentrations of atmospheric fine aerosols, i.e., fine particulate matter (PM\(_{1}\)), were observed at the remote island of Fukue, Nagasaki Prefecture, Japan (32.75° N, 128.68° E; see Supplementary Figure S1). The population on this island is approximately 40,000 and it is generally considered to have few emission sources. Aerosol chemical composition was measured using a quadrupole-type ACSM (Q-ACSM; Aerodyne Research Inc., Billerica, MA, USA). The mass concentrations of PM\(_{2.5}\) were obtained from an air-pollution monitoring station at Goto (located this island), which is the site of municipal government offices for Nagasaki prefecture. The chemical compositions and mass concentrations of atmospheric fine aerosols, i.e., fine particulate matter (PM\(_{1}\)), were observed at the remote island of Fukue, Nagasaki Prefecture, Japan (32.75° N, 128.68° E; see Supplementary Figure S1). The population on this island is approximately 40,000 and it is generally considered to have few emission sources. Aerosol chemical composition was measured using a quadrupole-type ACSM (Q-ACSM; Aerodyne Research Inc., Billerica, MA, USA). The mass concentrations of PM\(_{2.5}\) were obtained from an air-pollution monitoring station at Goto (located this island), which is the site of municipal government offices for Nagasaki prefecture. The chemical compositions and mass concentrations of atmospheric fine aerosols, i.e., fine particulate matter (PM\(_{1}\)), were observed at the remote island of Fukue, Nagasaki Prefecture, Japan (32.75° N, 128.68° E; see Supplementary Figure S1). The population on this island is approximately 40,000 and it is generally considered to have few emission sources. Aerosol chemical composition was measured using a quadrupole-type ACSM (Q-ACSM; Aerodyne Research Inc., Billerica, MA, USA). The mass concentrations of PM\(_{2.5}\) were obtained from an air-pollution monitoring station at Goto (located this island), which is the site of municipal government offices for Nagasaki prefecture.
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Author contributions
I.U. designed the study. I.U., S.I. and Z.W. coordinated all analyses. A.Y. and A.T. conducted ACSM measurements and analyzed the data for Fukue Island. M.H. converted the AEROS PM$_{2.5}$ data into gridded values. B.G.K. performed the data analysis for Korea. Y.Y. and K.Y. performed the overall data analysis. I.U. and Z.W. wrote the manuscript with valuable input from all coauthors.

Competing interests
The authors declare no competing interests.

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Correspondence and requests for materials should be addressed to I.U. or Z.W.

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