Source region attribution of PM$_{2.5}$ mass concentrations over Japan

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We estimated the contributions of source regions in East Asia to PM$_{2.5}$ mass concentrations over Japan for the year 2010 using a regional chemical transport model with an emission sensitivity approach. The horizontal distributions and temporal variations of PM$_{2.5}$ concentrations were generally well reproduced by the model. The relative contribution of China to the annual mean PM$_{2.5}$ concentration was estimated to be 50–60% in western Japan (from Kyushu to Kinki), and 40% in the Kanto area. Central north China (105°E–124°E, 34°N–42°N) made a particularly significant contribution, accounting for 20–40% of the annual mean PM$_{2.5}$ concentration for the whole of Japan. The contribution from foreign sources increased in spring, autumn, and winter in western Japan, and in spring in the Kanto area. The domestic contribution was estimated to be 20–50%. The sum of contributions from foreign anthropogenic sources was greater than the domestic pollution in each receptor region except the Kanto region. The results were obtained from sensitivity simulations with reduced anthropogenic emissions from each source region by 20%. The uncertainties in the source-receptor relationship for annual mean PM$_{2.5}$ concentrations were estimated to be within a few percent, from additional sensitivity simulations with different perturbations (a 20% increase and a 50% reduction). The model results suggest that regional-scale transport in East Asia has a significant impact on the attainment of the PM$_{2.5}$ environmental standard in Japan.

Keywords: PM$_{2.5}$, source-receptor relationship, chemical transport model, transboundary air pollution, aerosol

INTRODUCTION

Atmospheric aerosols have significant effects on air pollution and the Earth’s climate system. Aerosols alter the radiative budget by absorbing and scattering solar and thermal radiation and affecting cloud properties. Aerosol particles, also called particulate matter (PM), are considered to be one of the most important air pollutants. Of PM with a wide variation in size (from nanometers to micrometers in diameters), PM$_{2.5}$ (PM with a diameter of 2.5 μm or less) has received increasing attention since epidemiological studies have suggested links between its mass concentration and adverse health effects (U.S. Environmental Protection Agency (EPA), 2009). PM$_{2.5}$ contains both primary particles that are directly emitted to the atmosphere and secondary components that are formed by chemical reactions involving precursor gases such as SO$_2$, NO$_x$, and volatile organic compounds in the atmosphere. PM$_{2.5}$ is derived from anthropogenic or natural sources including fuel combustion in power plants and transport, biomass burning, vegetation, volcanoes, and dust.

In Japan, an environmental standard for PM$_{2.5}$ was newly established in September 2009, with an annual mean value of 15 μg m$^{-3}$ and a daily mean value of 35 μg m$^{-3}$. Since then, the number of monitoring sites for PM$_{2.5}$ has increased by the Japanese Ministry of the Environment (MOE) to understand the status of PM$_{2.5}$ pollution over Japan. Observations made by the MOE have indicated that an achievement ratio of the environmental standard determined at the ambient monitoring stations is about 30% for the fiscal years 2010 and 2011 (http://www.env.go.jp/air/osen/jokyo_h23/index.html). The monitoring stations at which PM$_{2.5}$ mass concentrations exceed the environmental standard are concentrated in the western part of Japan. In addition, Kanaya et al. (2010) reported that the PM$_{2.5}$ concentration at Fukue Island,
which is a remote island located in western Japan where the influence of local sources can be neglected, exceeded the standard value. These observations suggest that the levels of PM$_{2.5}$ pollution over Japan are influenced by PM$_{2.5}$ from source regions outside of Japan, especially the Asian continent, which is the main source region of global anthropogenic emissions of primary species and precursors of PM$_{2.5}$ (Streets et al., 2003). However, the relative importance of PM$_{2.5}$ transported from foreign source regions and domestic pollution on PM$_{2.5}$ concentrations over Japan is unclear. In the winter of 2013, a severe PM$_{2.5}$ pollution event occurred in Beijing and other areas in China, which has led to public concern in Japan about transboundary PM$_{2.5}$ pollution. To illustrate the extent to which PM$_{2.5}$ concentration in each receptor area is affected by domestic and various foreign sources, quantitative estimation of the contribution of PM$_{2.5}$ that can be attributed to each source region (i.e., source-receptor (S-R) relationship) using a chemical transport model is useful. Although there are several studies that have investigated the S-R relationship for PM$_{2.5}$ at the continental-scale (e.g., Asia, North America, and Europe) using global chemical transport models (e.g., Chin et al., 2007; Liu et al., 2009), they focused on intercontinental transport of PM$_{2.5}$ and thus did not address the regional-scale PM$_{2.5}$ pollution such as occurs in East Asia. The S-R relationship for PM$_{2.5}$ in the East Asian region has not yet been well examined compared with some other air pollutants such as acid deposition (Lin et al., 2008) and surface O$_3$ (Nagashima et al., 2010). Recently, several studies analyzed the transboundary pollution of aerosols using a chemical transport model with a sensitivity analysis and a tagged tracer method. Itahashi et al. (2012) quantified source contributions of sulfate aerosol from East Asia to Oki Island in Japan during air pollution episodes in the summer of 2005. Li et al. (2014) estimated the S-R relationship for PM$_{10}$ (PM with a diameter 10 $\mu$m or less) in Northeast Asia by a chemical transport model with the tracer tagged method. However, the S-R relationship has not been assessed for PM$_{2.5}$, which is of major concern in regional-scale air pollution in East Asia. Chatani et al. (2011) analyzed the sensitivity of transboundary transport from outside Japan to PM$_{2.5}$ concentrations in three major urban areas of Japan using a regional chemical transport model. However, because the monitoring sites where PM$_{2.5}$ concentrations exceed the environmental standard are distributed outside of these major urban areas, especially in the western part of Japan, an analysis for the whole of Japan is still required to clarify the entire picture of PM$_{2.5}$ pollution over Japan. In addition, to identify the source regions that contribute to air quality over Japan, it is useful to divide the East Asian region into different source areas, because foreign anthropogenic sources outside of Japan were treated as a single source in the previous study (Chatani et al., 2011). While Ikeda et al. (2014) investigated the contributions of PM$_{2.5}$ from several regions in East Asia, the receptor area was mainly targeted at Fukue Island located in western Japan. In this study, we estimated the contributions from seven source regions in East Asia to the PM$_{2.5}$ mass concentration over Japan for the year 2010 using a regional chemical transport model with an emission sensitivity approach.

METHODS

Model description
Meteorological fields to drive a chemical transport model were simulated by the Weather Research and Forecasting (WRF) model version 3.3.1 (Skamarock et al., 2008). The model domain, with the center 30°N, 115°E on the Lambert conformal projection, consisted of a 97×77 horizontal grid with a resolution of 80 km, covering the whole East Asian region (Fig. 1a), and had 37 levels in the vertical. The initial and boundary conditions were taken from the National Center for Environmental Pre-
combustion, industrial processes, agricultural activities, and so on, and considers the following air pollutants: SO\(_2\), NO\(_x\), CO, non-methane volatile organic compounds (NMVOCs), PM\(_{10}\), PM\(_{2.5}\), black carbon (BC), organic carbon (OC), NH\(_3\), and CH\(_4\). Because the target years of REAS version 2 are from 2000 to 2008, emissions for the latest available year (2008) were used for the CMAQ simulation of 2010. Biomass burning emissions were taken from the Global Fire Emission Database (GFED) version 3.1 for the year 2010. Biogenic emission data were taken from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2 (Guenther et al., 2006) for the year 2000. Volcanic emission data for SO\(_2\) were based on Streets et al. (2003) for the year 2000, but because the Miyakejima volcano was erupting during this period, the emission from it was modified to 1000 tons/day based on observations made by the Japan Meteorological Agency in 2010 (http://www.data.jma.go.jp/svd/vois/data/tokyo/320_Miyakejima/320_So2emission.htm). The method used to estimate the contribution of source regions

We used the emission sensitivity approach to estimate the contribution from each source region in East Asia to PM\(_{2.5}\) mass concentrations over Japan. In the emission sensitivity method, two kinds of simulations are conducted: a base run and a sensitivity run with emission perturbation. The contribution from each source region is calculated by taking the difference between the simulated PM\(_{2.5}\) concentrations of the base and each sensitivity experiment. Previous sensitivity studies have applied various magnitudes of perturbation on source emissions for sensitivity simulations. Chatani et al. (2011) quantified the sensitivity of transboundary transport of PM\(_{2.5}\) using a simulation in which anthropogenic emissions outside of Japan were reduced by 100%. Recent studies on source-receptor relationships for air pollutants used perturbations of 10–25% rather than a full removal to avoid a strong non-linear effect and to ensure a detectable response. Lin et al. (2008) estimated the S-R relationship for acid deposition in East Asia by sensitivity simulations with a 25% reduction. The model intercomparison studies by the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) applied a 20% reduction to quantify intercontinental ozone pollution (Fiore et al., 2009). In this study, we applied a 20% reduction in anthropogenic emissions from each source region for sensitivity simulations (Ikeda et al., 2014). All the emissions species in anthropogenic emissions were perturbed in sensitivity runs. To investigate the influence from different degrees of perturbation on the S-R relationship for PM\(_{2.5}\), we also performed sensitivity simulations with a 20% increase, and compared the results with those derived from sensitivity runs with a 20% reduction. Additionally, sensitivity simulations with a 50% reduction for
major source regions that had significant contributions to PM$_{2.5}$ concentrations over Japan were conducted to examine the uncertainties associated with the magnitude of the perturbations applied. Because the uncertainties in the S-R relationship for PM$_{2.5}$ due to the application of different perturbations to sensitivity simulations were very small, as discussed later, we have focused on the results derived from sensitivity simulations with a 20% reduction in the next chapter.

We divided East Asia into the seven source regions as shown in Fig. 1a: Japan (JPN), Korean Peninsula (KOR), Northeast China (CHN-NE), Central North China (CHN-CN), Central South China (CHN-CS), South China (CHN-S), and the rest of the model domain (OTH). In sensitivity simulations for these source regions, we perturbed the anthropogenic emissions of primary species and precursors of PM$_{2.5}$ from each region without changing the emissions from biomass burning, biogenic sources, and volcanoes. The contribution of natural sources (NAT) was quantified by a sensitivity simulation in which emissions from biomass burning, biogenic sources, and volcanoes were simultaneously perturbed in the whole model domain.

The contribution from a source $s$ to PM$_{2.5}$ mass concentration in a receptor area resulting from the ±20% sensitivity simulations, $A_{s,±20\%}$ was estimated as follows:

$$A_{s,±20\%} = 5 \times (C_{s,±20\%} - C_{s})$$

$$A_{s,±20\%} = 5 \times (C_{s,±20\%} - C_{s})$$

where $C_{s}$ is the concentration derived from the base run in which all of the emission sources were included, and $C_{s,±20\%}$ is the concentration derived from the sensitivity simulation with emissions from source $s$ perturbed by ±20%. The absolute contribution from each source was obtained by multiplying the difference of the simulated PM$_{2.5}$ between the base and sensitivity simulations by five, with the assumption of a linear relationship. The relative contributions ($R_{s,±20\%}$) were defined as the proportion of the contribution from source $s$ to the total value of contributions from all emission sources:

$$R_{s,±20\%} = \frac{A_{s,±20\%}}{\sum_{s=1}^{n} A_{s,±20\%}} \times 100.$$ 

To examine the extent of the influence due to the non-linearity to emission perturbations, we assessed the non-linear effect ($NL_{±20\%}$) which was defined based on the ratio of the sum of the contributions from all sources to the concentration derived from the base simulation:

$$NL_{±20\%} = \frac{\sum_{s=1}^{n} A_{s,±20\%}}{C_{\text{base}}} \times 100 - 100.$$ 

The area of Japan was divided into nine receptor areas (Fig. 1b), and the contributions from each source were estimated from the average concentrations within each receptor area. Model simulations were conducted for the period from December 1, 2009 to the end of December 2010, and we analyzed the results for the whole year of 2010.

**RESULTS AND DISCUSSION**

**Model evaluation**

The model results were evaluated by comparing them with observations made over Japan. We used PM$_{2.5}$ monitoring data obtained at Fukue Island by the Japan Agency for Marine-Earth Science and Technology (JAMSTEC) (Kanaya et al., 2010), Oki and Rishiri by the Acid Deposition Monitoring Network in East Asia (EANET), and 34 ambient monitoring stations operated by the MOE, Japan.

Figure 2 shows the horizontal distribution of annual mean PM$_{2.5}$ mass concentrations for the year 2010. Observed values at the ambient monitoring stations operated by the Japanese Ministry of the Environment are averaged for the fiscal year 2010.
tions being generally higher at sites in the western part of Japan (i.e., closer to the Asian continent), and decreasing toward the east of Japan. This trend in the spatial distribution of PM$_{2.5}$ concentrations is similar to that of observed particulate sulfate ($\text{SO}_4^{2-}$) concentrations reported by Aikawa et al. (2010), who analyzed $\text{SO}_4^{2-}$ concentrations measured at multiple sites over Japan during the period of 2003–2005. The model simulation successfully reproduced the spatial distribution in PM$_{2.5}$ concentrations, but absolute concentrations tended to be underestimated. It should be noted that observed PM$_{2.5}$ mass concentrations include an unidentified component, which is determined as the difference between total PM$_{2.5}$ and the sum of inorganic and carbonaceous components. According to a report by the Japanese Central Environmental Council, the contributions of unidentified components to total PM$_{2.5}$ were estimated to be about 20–40% based on measurements by the MOE over Japan for the period of 2004–2008 (https://www.env.go.jp/council/toshin/t07-h2102/01-1.pdf). The unidentified components may contain a certain amount of water (Kajino et al., 2006; Kanaya et al., 2010). It has been suggested that about 20% of the PM$_{2.5}$ concentration could consist of water according to the estimation using the E-AIM model (http://www.aim.env.uea.ac.uk/aim/aim.php) based on the measured chemical composition of PM$_{2.5}$ at Fukue in spring (Kanaya et al., 2010). Thus, the contribution of retained water in the measured PM$_{2.5}$ values may result in an un-

![Fig. 3. Seasonal variations of observed (gray line, left axis) and modeled (black line, right axis) monthly mean PM$_{2.5}$ mass concentrations in 2010.](image-url)
derestimation of PM$_{2.5}$ mass concentrations in the CMAQ simulation. Although dust may also be included as an unidentified component, its contribution to the annual average value is probably small as mentioned earlier. These biases may also be caused by the underestimation of OA in the CMAQ simulation (Chatani et al., 2011; Ikeda et al., 2014). Because the chemical composition and formation pathways of OA are not sufficiently understood, current chemical transport models including the CMAQ version 4.7.1 generally underestimate OA concentrations (Carlton et al., 2010; Hallquist et al., 2009). Carlton et al. (2010) reported that although the newly developed secondary organic aerosol module in CMAQ version 4.7.1 improves the performance compared with the previous mechanism, it is still difficult to accurately reproduce the concentration of OA. Uncertainties in the S-R relationship for PM$_{2.5}$ associated with the negative bias of the simulated concentrations will be examined in the next section.

Figure 3 shows a comparison of the seasonal variations in simulated monthly mean PM$_{2.5}$ concentrations with observations over Japan. Because PM$_{2.5}$ measurements at ambient monitoring stations by the MOE started in the fiscal year 2010, their results are shown from April. The vertical axis of the model (right axis) has been adjusted for comparison because the model underestimated the observations. At the sites located in the western part of Japan, i.e., from Kyushu to Kinki (Fukue, Oita, Fukuyama, Oki, Ino, and Sakai), the observed monthly mean PM$_{2.5}$ concentrations peaked in May and November, and decreased in September. Both peaks and the decrease in September were well simulated by the model. In Kanto (Yashio), the monthly average concentration increased slightly from spring to summer, and peaked in November after decreasing in September. In the Hokuriku and Tohoku areas (Imizu and Hachinohe), monthly mean PM$_{2.5}$ concentrations peaked during summer. In general, the regional differences in the seasonal variations of monthly average PM$_{2.5}$ concentrations over Japan were reproduced well by the model.

The model’s ability to reproduce day-to-day variations was evaluated by calculating correlation coefficients ($R$) of the daily mean PM$_{2.5}$ concentrations. The $R$ values were estimated to be 0.58–0.79 for the ambient monitoring stations and exceeded 0.70 for 60% of the sites. For the remote sites located in the western part of Japan, the model also reproduced daily variations of PM$_{2.5}$ concentrations well, with $R$ values of 0.68 and 0.80 for Fukue and Oki, respectively. These results suggest that the regional-scale transport that is important to estimate source contribution in East Asia was effectively simulated by the model.

<table>
<thead>
<tr>
<th>S/R</th>
<th>Contribution to annual mean concentrations from source regions (%)</th>
<th>Kyushu</th>
<th>Chugoku</th>
<th>Shikoku</th>
<th>Kinki</th>
<th>Hokuriku</th>
<th>Tohoku-Koshin</th>
<th>Kanto</th>
<th>Tohoku</th>
<th>Hokkaido</th>
</tr>
</thead>
<tbody>
<tr>
<td>JPN</td>
<td>21</td>
<td>25</td>
<td>23</td>
<td>36</td>
<td>33</td>
<td>46</td>
<td>51</td>
<td>30</td>
<td>19</td>
<td></td>
</tr>
<tr>
<td>KOR</td>
<td>10</td>
<td>11</td>
<td>8</td>
<td>6</td>
<td>6</td>
<td>3</td>
<td>0</td>
<td>4</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>CHN-NE</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>11</td>
<td>6</td>
<td>7</td>
<td>21</td>
<td>35</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CHN-CN</td>
<td>39</td>
<td>39</td>
<td>39</td>
<td>33</td>
<td>32</td>
<td>28</td>
<td>23</td>
<td>30</td>
<td>29</td>
<td></td>
</tr>
<tr>
<td>CHN-CS</td>
<td>15</td>
<td>13</td>
<td>13</td>
<td>11</td>
<td>11</td>
<td>10</td>
<td>8</td>
<td>8</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>CHN-S</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>OTH</td>
<td>4</td>
<td>4</td>
<td>4</td>
<td>4</td>
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<td>6</td>
<td></td>
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<tr>
<td>NAT</td>
<td>5</td>
<td>1</td>
<td>6</td>
<td>3</td>
<td>3</td>
<td>6</td>
<td>2</td>
<td>3</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 1. Relative contribution from source regions to receptors for annual mean PM$_{2.5}$ mass concentrations in 2010
anthropogenic sources.

The contribution of foreign anthropogenic sources, estimated as the sum of the contributions from the source regions except JPN, was largest in Kyushu close to the Asian continent, and smallest in Kanto. It should be noted that although the relative contribution of the foreign anthropogenic sources in Hokkaido was larger than in Kyushu, the absolute contribution was greater in Kyushu because the spatial distribution of PM$_{2.5}$ displayed a significant trend with higher concentrations in the western part of Japan. The relative contribution of the foreign anthropogenic sources was estimated to be 43–78% over Japan, which was greater than that of the domestic contribution (JPN) in most receptor areas, while domestic pollution was the main contributor (51%) in Kanto. Therefore, the model results suggest that the main PM$_{2.5}$ sources differ among the different areas of Japan. Among the foreign source regions, the contribution from China was dominant over Japan. The total contribution from the four source regions in China was estimated to be 39–61% in the receptor areas from Kyushu to Kanto. The atmospheric lifetime of PM$_{2.5}$ is approximately several days to one week (Textor et al., 2006), which is long enough for the particles to travel over several thousand kilometers (U.S. EPA, 2009). As a result, PM$_{2.5}$ originated from the East Asian continent (especially China), where anthropogenic emissions are quite large, can be transported to Japan located in the downwind region, and the model results suggest that its contribution has a significant influence on the air quality over Japan.

The S-R relationship for PM$_{2.5}$ resulting from sensitivity simulations with the 20% emission reduction were compared to those derived from sensitivity simulations with a 20% emission increase. The results from these perturbations were very similar to each other. The differences in the total contribution from foreign source regions between the −20% and +20% sensitivity simulations were less than 2% in every receptor area over Japan (Table 2). The differences between the total contributions from all the sources and the concentration of the base run ($C_{\text{base}}$) were estimated to be 5–12% (NL$_{-20\%}$). We also examined the non-linearity for individual species. The non-linear effects for SO$_4^{2–}$ and NH$_4^+$ were estimated to be −17~−11% and 8–17%, respectively. The non-linearity for NO$_3$ and secondary organic aerosol (SOA) were estimated to be 53–84% and 42–54%. Thus, non-linearity for total PM$_{2.5}$ could be largely caused by NO$_3^-$ and SOA. The non-linear effects determined from the 20% emission increases (NL$_{+20\%}$) were 11–15% in each receptor area, and were therefore similar to those of NL$_{-20\%}$. We further examined the dependence on the magnitude of the perturbation applied to sensitivity simulations. Sensitivity simulations with a 50% reduction in emissions from JPN or CHN-CN were conducted, and we estimated their contributions by taking the differences of simulated PM$_{2.5}$ between the base and perturbed simulations and multiplying them by two. The differences of the contribution from CHN-CN between the −20% and −50% perturbations were very small (within ±0.01 μg m$^{-3}$) in every receptor area (Table 2). The differences were also small for JPN at less than 0.16 μg m$^{-3}$ (corresponding to about 2% of the relative contribution). These results suggest that uncertainties in the S-R relationships for PM$_{2.5}$ caused by the different magnitudes of perturbation (i.e., −20%, +20%, and −50%) applied to the sensitivity simulations were within a few percent.

Uncertainties in the S-R relationship for PM$_{2.5}$ associated with the negative bias of the simulated concentrations were also examined. The comparison of each modeled chemical species with observations at Fukue indicated that the simulated SO$_4^{2–}$, NH$_4^+$, and EC, which are major components of PM$_{2.5}$, were very consistent with

<table>
<thead>
<tr>
<th>Source Region (SR)</th>
<th>Kyushu</th>
<th>Chugoku</th>
<th>Shikoku</th>
<th>Kinki</th>
<th>Hokuriku</th>
<th>Tokai-Koshin</th>
<th>Kanto</th>
<th>Tohoku</th>
<th>Hokkaido</th>
</tr>
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<tbody>
<tr>
<td>JPN</td>
<td>0.0</td>
<td>0.6</td>
<td>0.4</td>
<td>0.8</td>
<td>0.4</td>
<td>−0.2</td>
<td>−1.6</td>
<td>−0.3</td>
<td>−0.2</td>
</tr>
<tr>
<td>KOR</td>
<td>0.6</td>
<td>0.2</td>
<td>0.2</td>
<td>0.6</td>
<td>0.9</td>
<td>1.8</td>
<td>2.8</td>
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<td>0.7</td>
</tr>
<tr>
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<td>−0.1</td>
<td>−0.1</td>
<td>−0.2</td>
<td>−0.3</td>
<td>−0.3</td>
<td>−0.4</td>
<td>−0.5</td>
<td>−0.4</td>
<td>−0.3</td>
</tr>
<tr>
<td>CHN-CN</td>
<td>−0.6</td>
<td>−1.2</td>
<td>−0.9</td>
<td>−1.3</td>
<td>−1.6</td>
<td>−1.4</td>
<td>−1.0</td>
<td>−1.2</td>
<td>−0.8</td>
</tr>
<tr>
<td>CHN-CS</td>
<td>−0.1</td>
<td>−0.4</td>
<td>−0.1</td>
<td>−0.4</td>
<td>−0.6</td>
<td>−0.6</td>
<td>−0.6</td>
<td>−0.4</td>
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<td>CHN-S</td>
<td>−0.2</td>
<td>−0.1</td>
<td>−0.1</td>
<td>−0.1</td>
<td>−0.2</td>
<td>−0.1</td>
<td>−0.2</td>
<td>−0.1</td>
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<td>OTH</td>
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<td>0.4</td>
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<td>NAT</td>
<td>0.5</td>
<td>1.1</td>
<td>0.6</td>
<td>0.8</td>
<td>1.3</td>
<td>0.8</td>
<td>0.5</td>
<td>1.1</td>
<td>0.9</td>
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<td>JPN</td>
<td>−0.001</td>
<td>−0.089</td>
<td>−0.041</td>
<td>−0.126</td>
<td>−0.100</td>
<td>−0.160</td>
<td>−0.112</td>
<td>−0.043</td>
<td>0.004</td>
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<tr>
<td>CHN-CN</td>
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<td>0.014</td>
<td>0.014</td>
<td>0.007</td>
<td>0.006</td>
<td>0.002</td>
<td>−0.011</td>
<td>0.006</td>
<td>0.005</td>
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</table>

Source region attribution of PM$_{2.5}$ over Japan
the observed results, but OA was underestimated by the model. The mean bias of OA results corresponded to 12% of the observed PM$_{2.5}$ mass concentration (Ikeda et al., 2014). The underestimated OA and the unidentified components contained in the measured PM$_{2.5}$ were not reflected in the estimated S-R relationship for PM$_{2.5}$. If the model underestimates the PM$_{2.5}$ that originated from anthropogenic sources, the contribution from the source region to which the underestimated PM$_{2.5}$ fraction is attributed would have been underestimated. On the other hand, if the model underestimates the PM$_{2.5}$ that originated from natural sources, every value of the relative contribution from anthropogenic source regions would have been overestimated. For example, assuming that the relative bias due to OA and unidentified components corresponds to 30% of the observed PM$_{2.5}$ mass concentration in every receptor area and that the total amount of underestimated PM$_{2.5}$ should be attributed to the foreign source regions, the relative contribution of foreign anthropogenic sources would increase from 43–74% to 60–82% in Kyushu to Kanto. Conversely, if all of the underestimated PM$_{2.5}$ fraction is attributed to an origin other than the foreign source areas, the relative contribution of foreign anthropogenic sources would change to 30–52% in the same areas. Thus, it is suggested that uncertainties in the relative contribution of foreign anthropogenic sources associated with underestimated PM$_{2.5}$ are about ±15%, and it can be concluded that the impact of foreign sources is still significant over Japan including the Kanto area (30%) even in an extreme case where the total underestimated PM$_{2.5}$ fraction is assumed to originate from sources other than transboundary pollution.

Chatani et al. (2011) estimated the contribution of foreign anthropogenic sources to PM$_{2.5}$ concentrations in three major Japanese urban areas, using a sensitivity experiment with a 100% reduction in East Asian anthropogenic emissions except those from Japan. The sensitivity of transboundary pollution to the annual mean PM$_{2.5}$ concentration in Osaka–Hyogo, Aichi–Mie, and Shuto-ken areas was shown to be 48%, 41%, and 31%, respectively (Chatani et al., 2011). The corresponding contributions of transboundary pollution in the present study, i.e., the total of relative contributions from the six source regions other than JPN, were estimated to be 61%, 51%, and 43% in the Kinki, Tokai-Koshin, and Kanto receptor areas, where each major urban region considered by Chatani et al. (2011) is contained. Although direct comparison is not possible because of the differences in emission inventory, magnitude of emission perturbation, and the target year employed between these two studies, the results are almost similar, considering the studied uncertainty range. One reason for the somewhat larger values obtained in this study is likely to be the difference in the definition of receptor areas. This study uses wider receptor areas.
than those used by Chatani et al. (2011), who only considered the major urban areas and thus did not include surrounding regions in their receptor areas. In addition, the horizontal grid spacing of our model (80 km), to represent regional-scale features, was not fine enough to accurately represent the sub-grid variations of the emissions and the local-scale air pollution in urban areas.

Figure 4 shows seasonal variations of the contribution from each source region to the monthly mean PM$_{2.5}$ concentrations in Kyushu, Kinki, Kanto, and Tohoku resulting from the 20% reduction simulations. In Kyushu, the contributions from the foreign source regions, especially from CHN-CN and CHN-CS, increased in January, May, and November. They showed large variations even in the same season except during summer. In these seasons (i.e., spring, autumn, and winter), westerly winds are dominant in the mean wind field over East Asia, and this meteorological condition is basically favorable for the eastward transport of polluted air from the Asian continent to Japan. The monthly variations of the contributions from the foreign source regions are probably influenced by the frequency and intensity of transboundary transport events that occur in each month under the influence of these westerly winds (Ikeda et al., 2014). The seasonal behavior in Kinki was similar to that of Kyushu, with monthly elevations of the contributions from the source regions outside Japan observed in the same months. In Kanto, the contribution from CHN-CN had a spring peak in May, but, unlike in the western part of Japan, it did not increase during autumn and winter. This suggests that the eastward spread of the contributions from foreign source regions in autumn and winter was smaller than that during spring due to differences in transport patterns. While the fraction of PM$_{2.5}$ attributed to sources outside of Japan decreased during summer in Kyushu, Kinki, and Kanto, the contribution from CHN-CN increased in summer in Tohoku. This seasonal behavior was probably associated with the summer monsoon circulation, in which southerly winds intensify around the rim of the North Pacific High. These winds can prevent the transport of the polluted continental air mass to the western part of Japan and the Kanto area during summer. On the other hand, the contribution from CHN-CN can spread to relatively high latitude regions, i.e., the Sea of Japan, and may extend to northern Japan including the Tohoku area (not shown). As a result, in summer, the contribution from CHN-CN in the Tohoku area was larger than in western Japan and the Kanto area unlike other seasons. In general, domestic pollution was intensified in summer and decreased during winter.

CONCLUSIONS

We quantitatively estimated the relative contributions of anthropogenic sources from various regions in East Asia to PM$_{2.5}$ mass concentrations over Japan for the year 2010 using the regional chemical transport model WRF/CMAQ with emission sensitivity simulations. The model results were first evaluated by comparison with observational data over Japan. The observed annual mean PM$_{2.5}$ concentrations were generally higher in the western part of Japan, i.e., closer to the Asian continent, and decreased toward the east of Japan. The trend of the spatial distribution in observed PM$_{2.5}$ concentration was well reproduced by the model. The model tended to underestimate PM$_{2.5}$ mass concentrations, but regional differences in seasonal variations were well simulated by the model and the correlation coefficients of daily mean concentration were generally large, exceeding 0.70 for more than half of the sites.

The relative contribution from China to the annual mean PM$_{2.5}$ concentrations was estimated to be 40–60% from Kyushu to Kanto. The contribution of central north China was the most significant of all foreign source regions, accounting for 20–40% of the total across Japan. The contribution of transboundary pollution intensified in spring, autumn, and winter in the western part of Japan, and in spring in Kanto. While the sum of contributions from foreign anthropogenic sources was larger than that of the domestic contribution in most areas of Japan, domestic pollution was the main contributor to PM$_{2.5}$ pollution in Kanto. The model results suggested that foreign anthropogenic sources have a significant impact on the attainment of the environmental standard for PM$_{2.5}$ in Japan.

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REFERENCES


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