Comprehensive analyses of source sensitivities to and apportionments of PM$_{2.5}$ and ozone over Japan via multiple numerical techniques

Satoru Chatani$^1$, Hikari Shimadera$^2$, Syuichi Itahashi$^3$, Kazuyo Yamaji$^4$

$^1$National Institute for Environmental Studies, Tsukuba, Ibaraki 305-8506, Japan
$^2$Osaka University, Suita, Osaka 565-0871, Japan
$^3$Central Research Institute of Electric Power Industry, Abiko, Chiba 270-1194, Japan
$^4$Kobe University, Kobe, Hyogo 658-0022, Japan

Correspondence to: Satoru Chatani (chatani.satoru@nies.go.jp)

Supplementary material
Table S1: Annual total emission amounts (Mg/year) of each source group for the 2016 fiscal year in d02.

<table>
<thead>
<tr>
<th>Group</th>
<th>CO</th>
<th>SO₂</th>
<th>NOₓ</th>
<th>NH₃</th>
<th>NMVOC*¹</th>
<th>PM₂.₅</th>
<th>EC*²</th>
<th>OC*³</th>
</tr>
</thead>
<tbody>
<tr>
<td>s01</td>
<td>1,171,555</td>
<td>910</td>
<td>411,047</td>
<td>15,576</td>
<td>122,712</td>
<td>23,850</td>
<td>6,091</td>
<td>5,361</td>
</tr>
<tr>
<td>s02</td>
<td>59,862</td>
<td>279,102</td>
<td>682,708</td>
<td>0</td>
<td>20,593</td>
<td>55,988</td>
<td>10,766</td>
<td>16,045</td>
</tr>
<tr>
<td>s03</td>
<td>164,110</td>
<td>3,057</td>
<td>97,754</td>
<td>0</td>
<td>11,404</td>
<td>4,479</td>
<td>2,503</td>
<td>1,421</td>
</tr>
<tr>
<td>s04</td>
<td>1,230,684</td>
<td>356,649</td>
<td>761,388</td>
<td>0</td>
<td>226,562</td>
<td>19,196</td>
<td>1,661</td>
<td>3,459</td>
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<tr>
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<td>69,836</td>
<td>369</td>
<td>4,924</td>
<td>943</td>
<td>8,016</td>
<td>10,028</td>
<td>586</td>
<td>6,997</td>
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<td>840</td>
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<td>0</td>
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<td>1,770</td>
<td>190</td>
<td>619</td>
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<td>s07</td>
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<td>0</td>
<td>0</td>
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<td>0</td>
<td>0</td>
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<td>55,714</td>
<td>1,811</td>
<td>0</td>
<td>0</td>
<td>0</td>
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<tr>
<td>s09*⁴</td>
<td>404,322</td>
<td>1,534,513</td>
<td>58,700</td>
<td>21,732</td>
<td>3,954,064</td>
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</tr>
<tr>
<td>s10</td>
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<td>1,168,210</td>
<td>2,990,211</td>
<td>649,985</td>
<td>2,108,940</td>
<td>736,968</td>
<td>112,112</td>
<td>241,057</td>
</tr>
</tbody>
</table>

*¹ Non-methane volatile organic compound
*² Elemental carbon
*³ Organic carbon
*⁴ Lower values indicate amounts within Japan only
Table S2: Statistics of the model performance on the MDA8O3 and daily mean PM$_{2.5}$ concentrations for the entire 2016 fiscal year 2016 in the regions.

<table>
<thead>
<tr>
<th>Species</th>
<th>Region</th>
<th>Number$^1$</th>
<th>Obs.$^2$</th>
<th>Sim.$^3$</th>
<th>NMB$^4$</th>
<th>NME$^5$</th>
<th>R$^6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MDA8O3 (ppb)</td>
<td>JP</td>
<td>1150</td>
<td>42.7</td>
<td>46.0</td>
<td>7.79%</td>
<td>20.0%</td>
<td>0.860</td>
</tr>
<tr>
<td>KO</td>
<td>151</td>
<td>43.4</td>
<td>48.4</td>
<td>11.3%</td>
<td>24.0%</td>
<td>0.820</td>
<td></td>
</tr>
<tr>
<td>CS</td>
<td>143</td>
<td>44.4</td>
<td>46.8</td>
<td>5.27%</td>
<td>20.7%</td>
<td>0.834</td>
<td></td>
</tr>
<tr>
<td>KS</td>
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<td>46.2</td>
<td>7.14%</td>
<td>20.0%</td>
<td>0.868</td>
<td></td>
</tr>
<tr>
<td>TH</td>
<td>201</td>
<td>43.6</td>
<td>47.4</td>
<td>8.69%</td>
<td>18.9%</td>
<td>0.869</td>
<td></td>
</tr>
<tr>
<td>KK</td>
<td>369</td>
<td>41.5</td>
<td>44.3</td>
<td>6.94%</td>
<td>19.5%</td>
<td>0.872</td>
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</tr>
<tr>
<td>HT</td>
<td>110</td>
<td>39.4</td>
<td>44.1</td>
<td>11.7%</td>
<td>21.2%</td>
<td>0.797</td>
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<tr>
<td>OH</td>
<td>108</td>
<td>42.6</td>
<td>45.2</td>
<td>6.10%</td>
<td>20.7%</td>
<td>0.864</td>
<td></td>
</tr>
<tr>
<td>AM</td>
<td>75</td>
<td>42.6</td>
<td>45.6</td>
<td>6.89%</td>
<td>19.3%</td>
<td>0.874</td>
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<tr>
<td>ST</td>
<td>196</td>
<td>40.8</td>
<td>42.9</td>
<td>5.21%</td>
<td>19.7%</td>
<td>0.880</td>
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<tr>
<td>PM$_{2.5}$ (μg/m$^3$)</td>
<td>JP</td>
<td>820</td>
<td>11.9</td>
<td>7.62</td>
<td>-35.9%</td>
<td>41.6%</td>
<td>0.852</td>
</tr>
<tr>
<td>KO</td>
<td>127</td>
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<td>-27.2%</td>
<td>36.6%</td>
<td>0.860</td>
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<tr>
<td>CS</td>
<td>113</td>
<td>13.6</td>
<td>9.26</td>
<td>-30.3%</td>
<td>38.9%</td>
<td>0.853</td>
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<tr>
<td>KS</td>
<td>134</td>
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<td>7.77</td>
<td>-35.5%</td>
<td>39.5%</td>
<td>0.862</td>
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<tr>
<td>TH</td>
<td>132</td>
<td>10.7</td>
<td>6.62</td>
<td>-38.2%</td>
<td>42.0%</td>
<td>0.855</td>
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<tr>
<td>KK</td>
<td>243</td>
<td>11.3</td>
<td>6.48</td>
<td>-42.8%</td>
<td>46.2%</td>
<td>0.836</td>
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<tr>
<td>HT</td>
<td>71</td>
<td>9.01</td>
<td>5.41</td>
<td>-39.9%</td>
<td>46.0%</td>
<td>0.827</td>
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</tr>
<tr>
<td>OH</td>
<td>71</td>
<td>12.5</td>
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<td>-33.8%</td>
<td>38.2%</td>
<td>0.863</td>
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<tr>
<td>AM</td>
<td>43</td>
<td>11.6</td>
<td>7.40</td>
<td>-36.3%</td>
<td>40.0%</td>
<td>0.855</td>
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</tr>
<tr>
<td>ST</td>
<td>156</td>
<td>12.0</td>
<td>6.68</td>
<td>-44.5%</td>
<td>46.8%</td>
<td>0.839</td>
<td></td>
</tr>
<tr>
<td>SO$_4^{2-}$ (μg/m$^3$)</td>
<td>JP</td>
<td>154</td>
<td>2.73</td>
<td>2.64</td>
<td>-3.29%</td>
<td>40.9%</td>
<td>0.710</td>
</tr>
<tr>
<td>NO$_3^-$ (μg/m$^3$)</td>
<td>JP</td>
<td>154</td>
<td>0.641</td>
<td>1.01</td>
<td>-57.1%</td>
<td>121%</td>
<td>0.441</td>
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<tr>
<td>NH$_4^+$ (μg/m$^3$)</td>
<td>JP</td>
<td>154</td>
<td>1.11</td>
<td>1.08</td>
<td>-3.07%</td>
<td>41.1%</td>
<td>0.704</td>
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<tr>
<td>EC (μg/m$^3$)</td>
<td>JP</td>
<td>136</td>
<td>0.757</td>
<td>0.345</td>
<td>-54.4%</td>
<td>58.3%</td>
<td>0.477</td>
</tr>
<tr>
<td>OC (μg/m$^3$)</td>
<td>JP</td>
<td>151</td>
<td>2.58</td>
<td>0.958</td>
<td>-62.8%</td>
<td>66.0%</td>
<td>0.487</td>
</tr>
</tbody>
</table>

$^1$ Number of monitoring stations
$^2$ Observed values
$^3$ Simulated values
$^4$ Normalized Mean Bias
$^5$ Normalized Mean Error
$^6$ Correlation coefficient
Table S3: Source sensitivities to the annual mean ozone and PM$_{2.5}$ concentrations simulated in the regions. The upper table shows ratios (%) against the simulated concentrations. The lower table shows their normalized ratios (%).

(a1) O$_3$ (not normalized)

<table>
<thead>
<tr>
<th>Group</th>
<th>JP</th>
<th>KO</th>
<th>CS</th>
<th>KS</th>
<th>TH</th>
<th>KK</th>
<th>HT</th>
<th>OH</th>
<th>AM</th>
<th>ST</th>
</tr>
</thead>
<tbody>
<tr>
<td>s01</td>
<td>-0.8</td>
<td>-1.0</td>
<td>-0.5</td>
<td>-1.6</td>
<td>-1.1</td>
<td>-1.7</td>
<td>-0.1</td>
<td>-5.2</td>
<td>-5.9</td>
<td>-8.0</td>
</tr>
<tr>
<td>s02</td>
<td>0.6</td>
<td>0.7</td>
<td>-0.1</td>
<td>0.5</td>
<td>0.8</td>
<td>0.7</td>
<td>0.8</td>
<td>-1.7</td>
<td>-0.3</td>
<td>-0.5</td>
</tr>
<tr>
<td>s03</td>
<td>-0.2</td>
<td>-0.1</td>
<td>-0.1</td>
<td>-0.4</td>
<td>-0.3</td>
<td>-0.5</td>
<td>0.0</td>
<td>-1.7</td>
<td>-1.7</td>
<td>-2.5</td>
</tr>
<tr>
<td>s04</td>
<td>-0.6</td>
<td>-0.7</td>
<td>-1.2</td>
<td>-2.0</td>
<td>-0.6</td>
<td>-1.2</td>
<td>0.4</td>
<td>-8.3</td>
<td>-7.8</td>
<td>-9.6</td>
</tr>
<tr>
<td>s05</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>s06</td>
<td>-0.2</td>
<td>-0.1</td>
<td>-0.1</td>
<td>-0.3</td>
<td>-0.2</td>
<td>-0.3</td>
<td>-0.1</td>
<td>-1.0</td>
<td>-0.8</td>
<td>-1.6</td>
</tr>
<tr>
<td>s07</td>
<td>0.6</td>
<td>0.5</td>
<td>0.7</td>
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<td>1.1</td>
<td>0.2</td>
<td>1.5</td>
<td>1.8</td>
<td>2.3</td>
</tr>
<tr>
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<td>0.0</td>
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<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
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<td>0.0</td>
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</tr>
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</tr>
<tr>
<td>s10</td>
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<td>0.6</td>
<td>1.1</td>
<td>1.4</td>
<td>1.5</td>
<td>1.3</td>
<td>1.2</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
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<td>75.1</td>
<td>73.3</td>
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<td>-0.2</td>
<td>-0.2</td>
<td>-0.2</td>
<td>-0.2</td>
<td>-0.2</td>
<td>-0.1</td>
<td>-0.3</td>
<td>-0.3</td>
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<td>74.6</td>
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<td>76.3</td>
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(a2) O$_3$ (normalized)

<table>
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<tr>
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<th>CS</th>
<th>KS</th>
<th>TH</th>
<th>KK</th>
<th>HT</th>
<th>OH</th>
<th>AM</th>
<th>ST</th>
</tr>
</thead>
<tbody>
<tr>
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<td>-0.7</td>
<td>-2.1</td>
<td>-1.5</td>
<td>-2.3</td>
<td>-0.1</td>
<td>-7.6</td>
<td>-8.6</td>
<td>-11.7</td>
</tr>
<tr>
<td>s02</td>
<td>0.8</td>
<td>0.9</td>
<td>-0.1</td>
<td>0.7</td>
<td>1.1</td>
<td>0.9</td>
<td>1.0</td>
<td>-2.5</td>
<td>-0.4</td>
<td>-0.8</td>
</tr>
<tr>
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<td>-0.2</td>
<td>-0.2</td>
<td>-0.1</td>
<td>-0.6</td>
<td>-0.4</td>
<td>-0.6</td>
<td>0.0</td>
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</tr>
<tr>
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<td>0.4</td>
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<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
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</tr>
<tr>
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<td>-0.1</td>
<td>-0.4</td>
<td>-0.2</td>
<td>-0.4</td>
<td>-0.1</td>
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<td>1.9</td>
<td>1.5</td>
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<td>-0.3</td>
<td>-0.3</td>
<td>-0.2</td>
<td>-0.4</td>
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Table S3: Cont’d.

(b1) PM$_{2.5}$ (not normalized)

<table>
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<th>CS</th>
<th>KS</th>
<th>TH</th>
<th>KK</th>
<th>HT</th>
<th>OH</th>
<th>AM</th>
<th>ST</th>
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<td>7.8</td>
<td>7.5</td>
<td>6.2</td>
<td>4.0</td>
<td>10.1</td>
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</tr>
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<td>0.8</td>
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<td>0.5</td>
<td>1.5</td>
<td>1.8</td>
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</tr>
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Sum | 113.2| 114.0| 113.3| 113.4| 113.0| 117.6| 110.3| 114.5| 119.6| 123.4|

(b2) PM$_{2.5}$ (normalized)

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Figure S1: Comparisons of the observed and simulated monthly mean MDA8O3 and PM$_{2.5}$ concentrations at all stations in the regions. Markers and error bars represent mean values and standard deviations, respectively, of the daily concentrations at all monitoring stations for each month.
Figure S1: Cont'd
Figure S2: Scatter plots of the observed and simulated concentrations of PM$_{2.5}$ components during the monitoring campaigns of ambient concentrations of PM$_{2.5}$ components at all the locations throughout Japan in all four seasons. Regression lines are represented by red lines.
Figure S3: Source sensitivities to the annual mean concentrations of PM$_{2.5}$ components derived by BFM in the regions. Thick black lines represent the simulated concentrations.
Figure S4: Source sensitivities to the monthly mean concentrations of PM$_{2.5}$ components derived by BFM in entire Japan (JP) and ST. Thick black lines represent the simulated concentrations.
Figure S4: Cont’d
Figure S5: Apportionments derived by ISAM and sensitivities derived by BFM and HDDM of all source groups to the simulated concentrations of PM$_{2.5}$ components in JP and ST for the two target weeks in the four seasons.
Figure S5: Cont’d
Figure S6: Horizontal distributions of the apportionments and the sensitivities of s11 for the target two weeks of the spring.
Figure S7: Apportionments (left) and sensitivities (right) of NOX and VOC emissions of s01 derived by ISAM and BFM to the hourly ozone concentrations (shown by a line with markers) on July 25th in ST.
Figure S8: Horizontal distributions of the apportionments and sensitivities of the s01 NOx and VOC emissions to the ozone concentrations averaged for the two target weeks in summer.