Supplementary information for

Estimation of Secondary PM$_{2.5}$ in China and the United States using a Multi-Tracer Approach

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Table S1. Impacts of primary sulfate/nitrate emission uncertainty on the estimated secondary proportion of PM$_{2.5}$ in China.

<table>
<thead>
<tr>
<th>City</th>
<th>Change of sulfate/nitrate emission</th>
</tr>
</thead>
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<td></td>
<td>10%</td>
</tr>
<tr>
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<td>40.3</td>
</tr>
<tr>
<td>Tianjin</td>
<td>61.9</td>
</tr>
<tr>
<td>Shijiazhuang</td>
<td>44.8</td>
</tr>
<tr>
<td>Taiyuan</td>
<td>43.1</td>
</tr>
<tr>
<td>Hohhot</td>
<td>48.6</td>
</tr>
<tr>
<td>Shenyang</td>
<td>48.7</td>
</tr>
<tr>
<td>Changchun</td>
<td>47.9</td>
</tr>
<tr>
<td>Harbin</td>
<td>66.9</td>
</tr>
<tr>
<td>Shanghai</td>
<td>68.0</td>
</tr>
<tr>
<td>Nanjing</td>
<td>50.3</td>
</tr>
<tr>
<td>Hangzhou</td>
<td>45.6</td>
</tr>
<tr>
<td>Hefei</td>
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</tr>
<tr>
<td>Fuzhou</td>
<td>64.8</td>
</tr>
<tr>
<td>Nanchang</td>
<td>62.5</td>
</tr>
<tr>
<td>Ji'nan</td>
<td>54.6</td>
</tr>
<tr>
<td>Zhengzhou</td>
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</tr>
<tr>
<td>Changsha</td>
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<td>Guangzhou</td>
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</tr>
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<td>Haikou</td>
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<td>Lhasa</td>
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<tr>
<td>Urumqi</td>
<td>72.1</td>
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</table>

1 Based on the MEE observations in 2016.
Table S2. The comparison of two assumptions on weak correlation between secondary PM$_{2.5}$ and X-tracer in the secondary proportions of PM$_{2.5}$ (%) $^1$.

<table>
<thead>
<tr>
<th>City</th>
<th>$\delta 1$ $^2$</th>
<th>$\delta 2$ $^3$</th>
<th>$\delta 1 - \delta 2$</th>
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<td>0.5</td>
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<td>42.8</td>
<td>2.0</td>
</tr>
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<td>43.1</td>
<td>40.9</td>
<td>2.2</td>
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<td>48.6</td>
<td>46.8</td>
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<td>47.0</td>
<td>1.7</td>
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<td>1.6</td>
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<td>66.9</td>
<td>68.3</td>
<td>-1.4</td>
</tr>
<tr>
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<td>68.0</td>
<td>67.8</td>
<td>0.2</td>
</tr>
<tr>
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<td>50.3</td>
<td>47.6</td>
<td>2.7</td>
</tr>
<tr>
<td>Hangzhou</td>
<td>45.6</td>
<td>43.4</td>
<td>2.2</td>
</tr>
<tr>
<td>Hefei</td>
<td>65.4</td>
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<td>0.3</td>
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<td>52.8</td>
<td>1.9</td>
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<td>60.4</td>
<td>1.1</td>
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<td>Changsha</td>
<td>65.9</td>
<td>65.7</td>
<td>0.2</td>
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<td>-0.4</td>
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<td>61.9</td>
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<td>72.1</td>
<td>73.3</td>
<td>-1.2</td>
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</table>

$^1$ Based on the MEE observations in 2016.

$^2$ $\delta 1$ is mean value of the estimated interval.

$^3$ $\delta 2$ is the specific value when $r=0$. 
Table S3. List of 31 populous cities and 19 regional background cities and the corresponding averaged PM$_{2.5}$ concentrations ($\mu$g m$^{-3}$) in China during the studying period.

<table>
<thead>
<tr>
<th>Province</th>
<th>Populous city</th>
<th>PM$_{2.5}$</th>
<th>Regional background city</th>
<th>PM$_{2.5}$</th>
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<td>Taiyuan</td>
<td>62.8</td>
<td>-</td>
<td>-</td>
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<td>Xilingol</td>
<td>16.2</td>
</tr>
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<td>Shenyang</td>
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<td>-</td>
<td>-</td>
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<td>-</td>
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<td>-</td>
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<td>-</td>
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<td>-</td>
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<td>Lanzhou</td>
<td>50.1</td>
<td>Jiayuguan</td>
<td>30.9</td>
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<td>Xining</td>
<td>49.0</td>
<td>Yushu</td>
<td>18.0</td>
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<td>Yin chuan</td>
<td>44.7</td>
<td>Guyuan</td>
<td>35.6</td>
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<td>Xinjiang</td>
<td>Urumqi</td>
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<td>Altay</td>
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<td>Period</td>
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<td>45.5</td>
</tr>
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</tr>
<tr>
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<td>2012 Winter</td>
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<td>11.6 13.2 5.6 8.5</td>
<td>55.6%</td>
</tr>
<tr>
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</tr>
<tr>
<td></td>
<td>Jun 2014 – Jul 2014</td>
<td>56</td>
<td>6.7 2.8 2.1 2.9</td>
<td>25.9%</td>
</tr>
<tr>
<td></td>
<td>2013 Winter</td>
<td>91</td>
<td>10.8 12.4 7.5 21.8&lt;sup&gt;b&lt;/sup&gt;</td>
<td>57.7%</td>
</tr>
<tr>
<td></td>
<td>Dec 2014 – Jan 2015</td>
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<td>18.3 25.4 14.4 14.1&lt;sup&gt;b&lt;/sup&gt;</td>
<td>70.1%</td>
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<sup>a</sup> Indicates data from a different source. <sup>b</sup> Indicates a different measurement method.
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1 SOA = 0.5*OM, OM = f * OC. Default f is 1.2. In case of a, b and c, the f is 1.8, 1.6 and 1.4 respectively.
Table S5. Impacts of anthropogenic emission uncertainty on the estimated secondary proportion of PM$_{2.5}$ $^1$ in China.

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<th>City</th>
<th>Secondary proportion of PM$_{2.5}$</th>
<th>Change of secondary proportion of PM$_{2.5}$ $^a$</th>
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$^1$ Based on the MEE observations in 2016.
Figure S1. An illustration for scanning the aim interval (green shadow) in the MTEA approach.
Figure S2. The spatial distribution of (a-b) NO\textsubscript{2}, (d-e) CO and (g-h) maximum daily 8-h average O\textsubscript{3} (MDA8) mass concentrations before (01~23 Jan 2020) and during (23-Jan ~ 17-Feb 20) COVID-19 national lockdown. The right panel (c, f, and g) indicates relative change, i.e. (post-lockdown – pre-lockdown)/pre-lockdown.
Figure S3. The nationwide correlation between PM versus O₃. Blue bars denote the correlation coefficients between SPM and O₃. The red dots indicate the ratios of the correlation coefficient of PPM vs O₃ to that of SPM vs O₃.