Temporospatial Variation, Chemical Composition, and Source Resolution of PM$_{2.5}$ in the Southeastern Taiwan Island

(Supporting Information)

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A. PM$_{2.5}$ Sampling

During the sampling periods, PM$_{2.5}$ samples were collected by PM$_{2.5}$ samplers (BGI, PQ-200) in an air flowrate of 16.67 L min$^{-1}$ at the two remote sites. The dates of PM$_{2.5}$ sampling were September 17-23, 2020 (Fall), January 10-16, 2021 (Winter), April 12-18, 2021 (Spring), and August 11-17, 2021 (Summer). 24-hr PM$_{2.5}$ samples were simultaneously collected at the two remote sites for consecutive seven days in each season. Quartz filters (Advantec, QR-100, 47 mm) were used to collect PM$_{2.5}$ samples. Prior to weighing, the PM$_{2.5}$ filters must be conditioned in a desiccator with the relative humidity (RH) of 40±5% and the temperature of 20±5°C for at least 48-hr to avoid the potential interference of moisture. An analytical microbalance (Sartorius, MSA6.6S) with the precision of 10$^{-6}$ g was used to precisely weigh the filters. The mass difference of each PM$_{2.5}$ filter before and after sampling was calculated as the mass of collected PM$_{2.5}$. The concentration of PM$_{2.5}$ was then determined by dividing the mass of collected PM$_{2.5}$ with the total amount of air sampling volume.

B. Chemical Analytical Methods

Firstly, we tailor half of the PM$_{2.5}$ filter for analyzing the water-soluble ions and put it into a 50-mL PE bottle pouring 30 ml deionized water (D.I. H$_2$O) with conductivity >18 MΩ for 120 min in an ultrasonic vibrator (Branson, 5510) in order to extract the solid-phase ionic species of PM$_{2.5}$ into the liquid extraction solution. An ion chromatography (Thermo Fisher, Dionex, ICS-1100) was used to analyze the concentrations of major anions (F$^-$, Cl$^-$, Br$^-$, NO$_3^-$, SO$_4^{2-}$) and major cations (Na$^+$, NH$_4^+$, K$^+$, Mg$^{2+}$, Ca$^{2+}$) with 3.6 mM Na$_2$CO$_3$/4.5 mM NaHCO$_3$ solution and 5 mM methane sulfonic acid as eluent, respectively.

One-quarter of the PM$_{2.5}$ filter was tailored for analyzing the metallic elements, which was initially digested in 30 mL with an acidic mixture (HNO$_3$: HClO$_3$ = 3:7) at 180°C for
40 min in a microwave digester (PreeKem, TOPEX+). The digested solution was then
diluted to 50 ml with D.I. H₂O after microwave digestion. An inductively coupled
plasma-atomic emission spectrometry (ICP-AES) (Perkin Elmer, Model Optima 2000DV)
was used to analyze the concentrations of 17 metallic elements including Ba, Cr, Mn, Ni, Zn,
Ca, Cd, Fe, Pb, Mg, K, Ti, Al, As, V, Cu, and Sb.

Further tailoring the PM₂.₅ filters into two identical portions of one eighth was used to
analyze the carbonaceous contents in PM₂.₅ with an elemental analyzer (EA) (Elementar,
Vario MICRO cube). One-eighth of the PM₂.₅ filter was analyzed posterior to preheat at
350°C in a muffle furnace for 30 min to expel the organic carbon (OC) fraction. Another
eighth of the PM₂.₅ filter was analyzed without preheating for respectively characterizing
the elemental carbon (EC) and total carbon (TC). After the analytical process, organic
carbon (OC) was determined by extracting EC from TC.

C. Enrichment Factor

The equation of EF was expressed by Eq. (S1)

\[ EF = \frac{(Tr/Ref)_{PM}}{(Tr/Ref)_{crust}} \]  \hspace{1cm} (S1)

where \( Tr \) is the concentrations of metallic elements in PM₂.₅ emitted from the pollution
sources; \( Ref \) is the concentration of a reference element in PM₂.₅ emitted from the crustal.
Iron (Fe) is a main crustal element and conserved element which is often used as the
reference element for calculating EF. Therefore, this study used Fe as the reference element.
As EF>10, it indicates that the metallic element is mostly contributed by anthropogenic
sources; As EF<1, it indicates that the metallic element is originated mainly from the crust.
As 1<EF<10, it indicates that the metallic element may be originated from both combined
crustal and anthropogenic sources.
D. Backward Trajectory Simulation

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model developed by the Air Resources Laboratory of National Oceanic and Atmospheric Administration (NOAA) has been widely applied to determine either the forward or backward trajectories of air parcels in a large space scale. To determine the transport routes of air pollutants, this study simulated the past 72-hr backward trajectories of air masses reaching the target sites at the height of 100 m by HYSPLIT backward trajectories.

E. Chemical Mass Balance (CMB) Receptor Model

The source apportionment of PM$_{2.5}$ sampled at the southeastern sea aeras of Taiwan Island was resolved by using a Chemical Mass Balance (CMB, Version 8.0) receptor model which was developed by Desert Research Institute (DRI) based on the principle of mass conservation. It is essential to measure the PM$_{2.5}$ of comprehensive knowledge of the physical and chemical characteristics to identify the potential sources and quantify their contributions to PM$_{2.5}$ at each receptor site. The equation between the mass concentrations of chemical species measured at the receptor site and those emitted from the potential sources can be expressed by Eq. (S2).

\[ C_i = \sum_{j=1}^{P} \alpha_{ij} F_{ij} S_j \]  

(S2)

where $C_i$ is the concentration of species i measured at the receptor site (µg m$^{-3}$); $\alpha_{ij}$ is the coefficient of species i generated or removed during the transport process; $F_{ij}$ is the mass fraction of species i in the chemical profile of the source j (%); $S_j$ is the mass concentration of all species at the receptor site assigned to the source j (µg m$^{-3}$).
Table Captions

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**Table S1. Geographical location and environmental description of two remote sites.**

<table>
<thead>
<tr>
<th>Sampling Sites</th>
<th>Longitude/Latitude</th>
<th>Environmental Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Green Island (GI)</td>
<td>121°50′04″E 22°67′51″N</td>
<td>Green Island is an offshore island located at the western Pacific Ocean, which is about 33 km away from Taitung City. It is the fourth largest offshore island in Taiwan. The local resident population is about 4,000, but the number of tourists during the tourist season (June-November) is as high as 97,160.</td>
</tr>
<tr>
<td>Kenting Peninsula (KP)</td>
<td>120°85′26″E 21°90′24″N</td>
<td>Kenting Peninsula is located at the southernmost tip of the Taiwan Island. It is surrounded by the sea on three sides, the western Pacific Ocean to the east, the Bashi Strait to the south, and the Taiwan Strait to the west. It is the first National Park in Taiwan and a famous tourist destination in the south Taiwan. The population of local residents is about 1,500, but the number of tourists during the tourist season (June-November) is as high as 79,960.</td>
</tr>
</tbody>
</table>
Table S2  Seasonal variation of meteorological data measured at the GI and KP during the sampling period.

<table>
<thead>
<tr>
<th></th>
<th>Site</th>
<th>Air Temperature (°C)</th>
<th>Relative Humidity (%)</th>
<th>Wind Speed (m/s)</th>
<th>Prevailing Wind Direction</th>
<th>Precipitation (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fall</td>
<td>GR</td>
<td>27.6±0.8</td>
<td>84.9±4.5</td>
<td>4.1±2.6</td>
<td>NNE</td>
<td>44.5</td>
</tr>
<tr>
<td></td>
<td>KT</td>
<td>28.1±0.5</td>
<td>87±3.8</td>
<td>1.9±0.6</td>
<td>E</td>
<td>30.5</td>
</tr>
<tr>
<td>Winter</td>
<td>GR</td>
<td>16.7±2.2</td>
<td>75.1±7.2</td>
<td>8.0±2.0</td>
<td>NNE</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>KT</td>
<td>18.6±1.8</td>
<td>73±5.9</td>
<td>3.9±0.7</td>
<td>NE</td>
<td>1.5</td>
</tr>
<tr>
<td>Spring</td>
<td>GR</td>
<td>23.3±0.7</td>
<td>87.1±7.6</td>
<td>5.6±3.6</td>
<td>NNE</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>KT</td>
<td>25.7±0.8</td>
<td>77.6±4.9</td>
<td>4±2</td>
<td>NEE</td>
<td>0</td>
</tr>
<tr>
<td>Summer</td>
<td>GR</td>
<td>27.7±0.7</td>
<td>89.7±3.4</td>
<td>2.80±1.0</td>
<td>SE</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>KT</td>
<td>27.9±0.5</td>
<td>91.1±2</td>
<td>1.2±1</td>
<td>SW</td>
<td>46</td>
</tr>
</tbody>
</table>
Table S3. Correlation between PM$_{2.5}$ and Cl$^-$ Deficit at two remote islands.

<table>
<thead>
<tr>
<th>Seasons</th>
<th>Sites</th>
<th>Correlation between PM$_{2.5}$ and Cl$^-$ Deficit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fall</td>
<td>GI</td>
<td>0.54</td>
</tr>
<tr>
<td></td>
<td>KP</td>
<td>0.63</td>
</tr>
<tr>
<td>Winter</td>
<td>GI</td>
<td>0.81</td>
</tr>
<tr>
<td></td>
<td>KP</td>
<td>0.84</td>
</tr>
<tr>
<td>Spring</td>
<td>GI</td>
<td>0.63</td>
</tr>
<tr>
<td></td>
<td>KP</td>
<td>0.59</td>
</tr>
<tr>
<td>Summer</td>
<td>GI</td>
<td>0.21</td>
</tr>
<tr>
<td></td>
<td>KP</td>
<td>0.13</td>
</tr>
</tbody>
</table>

*Red:* significant correlation coefficient; *Yellow:* moderated correlation coefficient; *Blue:* poor correlation coefficient; *Green:* uncorrelated coefficient.
Table S4. Correlation analysis of PM$_{2.5}$ and its chemical species at the sites GI and KP by using a pair T-test.

<table>
<thead>
<tr>
<th>Chemical Species</th>
<th>Fall</th>
<th>Winter</th>
<th>Spring</th>
<th>Summer</th>
</tr>
</thead>
<tbody>
<tr>
<td>PM$_{2.5}$</td>
<td>0.315</td>
<td>0.078</td>
<td>0.031</td>
<td>0.033</td>
</tr>
<tr>
<td>Cl$^-$</td>
<td>0.656</td>
<td>0.190</td>
<td>0.674</td>
<td>0.054</td>
</tr>
<tr>
<td>NO$_3^-$</td>
<td>0.135</td>
<td>0.609</td>
<td>0.835</td>
<td>0.001</td>
</tr>
<tr>
<td>SO$_4^{2-}$</td>
<td>0.332</td>
<td>0.008</td>
<td>0.000</td>
<td>0.442</td>
</tr>
<tr>
<td>Na$^+$</td>
<td>0.637</td>
<td>0.080</td>
<td>0.383</td>
<td>0.044</td>
</tr>
<tr>
<td>NH$_4^+$</td>
<td>0.759</td>
<td>0.016</td>
<td>0.729</td>
<td>0.024</td>
</tr>
<tr>
<td>K$^+$</td>
<td>0.002</td>
<td>0.022</td>
<td>0.172</td>
<td>0.011</td>
</tr>
<tr>
<td>Mg$^{2+}$</td>
<td>0.009</td>
<td>0.879</td>
<td>0.012</td>
<td>0.002</td>
</tr>
<tr>
<td>Ca$^{2+}$</td>
<td>0.039</td>
<td>0.859</td>
<td>0.063</td>
<td>0.590</td>
</tr>
<tr>
<td>Mg</td>
<td>0.025</td>
<td>0.614</td>
<td>0.005</td>
<td>0.000</td>
</tr>
<tr>
<td>Al</td>
<td>0.194</td>
<td>0.734</td>
<td>0.083</td>
<td>0.004</td>
</tr>
<tr>
<td>Ca</td>
<td>0.083</td>
<td>0.618</td>
<td>0.046</td>
<td>0.453</td>
</tr>
<tr>
<td>Ti</td>
<td>0.769</td>
<td>0.000</td>
<td>0.000</td>
<td>0.006</td>
</tr>
<tr>
<td>V</td>
<td>0.709</td>
<td>0.064</td>
<td>0.374</td>
<td>0.924</td>
</tr>
<tr>
<td>Mn</td>
<td>0.433</td>
<td>0.037</td>
<td>0.329</td>
<td>0.008</td>
</tr>
<tr>
<td>Fe</td>
<td>0.391</td>
<td>0.477</td>
<td>0.770</td>
<td>0.354</td>
</tr>
<tr>
<td>Ni</td>
<td>0.791</td>
<td>0.023</td>
<td>0.955</td>
<td>0.099</td>
</tr>
<tr>
<td>Zn</td>
<td>0.269</td>
<td>0.809</td>
<td>0.078</td>
<td>0.000</td>
</tr>
<tr>
<td>Pb</td>
<td>0.175</td>
<td>0.115</td>
<td>0.729</td>
<td>0.074</td>
</tr>
<tr>
<td>Cr</td>
<td>0.367</td>
<td>0.053</td>
<td>0.588</td>
<td>0.000</td>
</tr>
<tr>
<td>Cu</td>
<td>0.940</td>
<td>0.009</td>
<td>0.224</td>
<td>0.278</td>
</tr>
<tr>
<td>Cd</td>
<td>0.350</td>
<td>0.059</td>
<td>0.121</td>
<td>0.181</td>
</tr>
<tr>
<td>K</td>
<td>0.284</td>
<td>0.671</td>
<td>0.210</td>
<td>0.003</td>
</tr>
<tr>
<td>OC</td>
<td>0.255</td>
<td>0.206</td>
<td>0.042</td>
<td>0.027</td>
</tr>
<tr>
<td>EC</td>
<td>0.281</td>
<td>0.184</td>
<td>0.034</td>
<td>0.038</td>
</tr>
<tr>
<td>TC</td>
<td>0.259</td>
<td>0.199</td>
<td>0.034</td>
<td>0.033</td>
</tr>
</tbody>
</table>

Red: p-value < 0.01, Strongly significant; Brown: p-value < 0.05, significant; Gray: p-value > 0.05, insignificant.
Table S5. Spatiotemporal variation of PM$_{2.5}$’s source apportionment during the sampling periods.

<table>
<thead>
<tr>
<th>Sampling Sites</th>
<th>Sources</th>
<th>Fall</th>
<th>Winter</th>
<th>Spring</th>
<th>Summer</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>GI</td>
<td>KP</td>
<td>GI</td>
<td>KP</td>
<td>GI</td>
</tr>
<tr>
<td>Incinerators</td>
<td>0.23 3.1</td>
<td>0.18 3.2</td>
<td>0.49 4.7</td>
<td>0.60 4.6</td>
<td>0.49 3.9</td>
</tr>
<tr>
<td>Petrochemical Industry</td>
<td>0.17 2.3</td>
<td>0.12 2.1</td>
<td>0.34 3.2</td>
<td>0.31 2.4</td>
<td>0.43 3.4</td>
</tr>
<tr>
<td>Steel Plants</td>
<td>0.10 1.3</td>
<td>0.06 1.1</td>
<td>0.20 1.9</td>
<td>0.28 2.1</td>
<td>0.15 1.2</td>
</tr>
<tr>
<td>Industrial Boilers (Oil-fired)</td>
<td>0.92 12.4</td>
<td>0.69 12.3</td>
<td>0.94 9.0</td>
<td>1.63 12.5</td>
<td>1.46 11.5</td>
</tr>
<tr>
<td>Industrial Boilers (Coal)</td>
<td>0.01 0.2</td>
<td>0.01 0.2</td>
<td>0.14 1.4</td>
<td>0.28 2.1</td>
<td>0.24 1.9</td>
</tr>
<tr>
<td>Fugitive Dust</td>
<td>1.46 19.6</td>
<td>1.08 19.2</td>
<td>2.17 20.7</td>
<td>2.70 20.6</td>
<td>2.67 21.0</td>
</tr>
<tr>
<td>Mobile Sources</td>
<td>0.74 10.0</td>
<td>0.46 8.2</td>
<td>0.80 7.6</td>
<td>0.85 6.5</td>
<td>0.97 7.6</td>
</tr>
<tr>
<td>Biomass Burning</td>
<td>0.29 3.9</td>
<td>0.21 3.7</td>
<td>0.57 5.4</td>
<td>0.76 5.8</td>
<td>0.61 4.8</td>
</tr>
<tr>
<td>Sea Salts</td>
<td>1.53 20.5</td>
<td>1.08 19.1</td>
<td>2.23 21.2</td>
<td>2.46 18.8</td>
<td>2.40 18.9</td>
</tr>
<tr>
<td>Cement Industry</td>
<td>0.00 0.0</td>
<td>0.00 0.0</td>
<td>0.00 0.0</td>
<td>0.00 0.0</td>
<td>0.00 0.0</td>
</tr>
<tr>
<td>Secondary Sulfate</td>
<td>0.70 9.4</td>
<td>0.53 9.3</td>
<td>1.06 10.1</td>
<td>1.28 9.8</td>
<td>1.07 8.4</td>
</tr>
<tr>
<td>Secondary Nitrate</td>
<td>0.17 2.3</td>
<td>0.17 3.0</td>
<td>0.46 4.4</td>
<td>0.50 3.9</td>
<td>0.50 3.9</td>
</tr>
<tr>
<td>Secondary Organic Carbon</td>
<td>0.14 1.9</td>
<td>0.07 1.3</td>
<td>0.27 2.6</td>
<td>0.25 1.9</td>
<td>0.24 1.9</td>
</tr>
<tr>
<td>Fishing Boats</td>
<td>0.20 2.7</td>
<td>0.22 3.9</td>
<td>0.22 2.1</td>
<td>0.60 4.6</td>
<td>0.31 2.4</td>
</tr>
<tr>
<td>Percent Mass</td>
<td>89.5%</td>
<td>86.3%</td>
<td>94.0%</td>
<td>95.4%</td>
<td>90.6%</td>
</tr>
<tr>
<td>$\chi^2$</td>
<td>1.28</td>
<td>1.22</td>
<td>1.37</td>
<td>1.40</td>
<td>1.30</td>
</tr>
<tr>
<td>R$^2$</td>
<td>0.88</td>
<td>0.85</td>
<td>0.93</td>
<td>0.94</td>
<td>0.90</td>
</tr>
<tr>
<td>t statistic</td>
<td>12.95</td>
<td>13.18</td>
<td>14.08</td>
<td>14.95</td>
<td>13.94</td>
</tr>
</tbody>
</table>
Table S6. Contribution of CBT to PM$_{2.5}$ at the Green Island and the Kenting Peninsula.

<table>
<thead>
<tr>
<th>Seasons</th>
<th>Fall</th>
<th>Winter</th>
<th>Spring</th>
<th>Summer</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>GI</td>
<td>KP</td>
<td>GI</td>
<td>KP</td>
</tr>
<tr>
<td>PM$_{2.5}$</td>
<td>7.47</td>
<td>5.66</td>
<td>10.53</td>
<td>13.11</td>
</tr>
<tr>
<td>CBT</td>
<td>3.83</td>
<td>3.02</td>
<td>6.89</td>
<td>10.44</td>
</tr>
<tr>
<td>CBT/PM$_{2.5}$</td>
<td>51.2%</td>
<td>53.3%</td>
<td>65.5%</td>
<td>79.7%</td>
</tr>
</tbody>
</table>

Units: $\mu$g/m$^3$; GI: Green Island; KP: Kenting Peninsula; CBT: cross-boundary transport
**Table S7.** Recovery rates of metallic elements in PM$_{2.5}$ at the two remote islands in the SCS.

<table>
<thead>
<tr>
<th>Metals</th>
<th>Cr</th>
<th>Ni</th>
<th>As</th>
<th>Cd</th>
<th>Pb</th>
<th>Ti</th>
<th>Al</th>
<th>Fe</th>
<th>Mn</th>
<th>Zn</th>
<th>Ca</th>
<th>K</th>
<th>Mg</th>
<th>V</th>
<th>Cu</th>
<th>Ba</th>
<th>Sb</th>
</tr>
</thead>
<tbody>
<tr>
<td>MDL</td>
<td>0.017</td>
<td>0.005</td>
<td>0.017</td>
<td>0.002</td>
<td>0.037</td>
<td>0.011</td>
<td>0.062</td>
<td>0.011</td>
<td>0.020</td>
<td>0.005</td>
<td>0.035</td>
<td>0.062</td>
<td>0.036</td>
<td>0.009</td>
<td>0.006</td>
<td>0.0008</td>
<td>0.002</td>
</tr>
<tr>
<td>R$^2$</td>
<td>0.9998</td>
<td>0.9985</td>
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MDL: method detection limit (mg/L); R$^2$: determination coefficient; RR: recovery rate
**Table S8.** Source profile applied for resolving the source apportionment of PM$_{2.5}$ by using CMB receptor model.

<table>
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<tr>
<th>Codes</th>
<th>Abbreviates</th>
<th>Emission Sources</th>
<th>Literatures</th>
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<td>PBPRI1</td>
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<td>SCT023</td>
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<td>Chen et al., 1997</td>
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**Figure Captions**

**Fig. S1.** Location of PM$_{2.5}$ sampling sites at the Green Island and the Kenting Peninsula in the southeastern sea areas of Taiwan Island.

**Fig. S2.** Source apportionment of PM$_{2.5}$ at two remote sites during the sampling periods resolved by CMB receptor model.

**Fig. S3.** The superimposition of backward trajectories starting from (a) the Green Island and (b) the Kenting Peninsula on the regional fire maps of East Asia in four seasons.

**Fig. S4.** Spatial distribution of marine PM$_{2.5}$ at the island and coastal areas in East Asia.

**Fig. S5** The trajectory map of ships navigated in the seas of East Asia.
Fig. S1. Location of PM$_{2.5}$ sampling sites at the Green Island and the Kenting Peninsula in the southeastern sea areas of Taiwan Island.
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Fig. S5 The trajectory map of ships navigated in the seas of East Asia.