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

Chung-Shin Yuan$^{1,2,*}$, Jun-Hao Ceng$^1$, Po-Hsuan Yen$^1$, Kuan-Chen Chiang$^1$, Yu-Lun Tseng$^1$, Kwok-Wai Wong$^1$, Ming-Shiou Jeng$^{3,4}$

$^1$ Institute of Environmental Engineering, National Sun Yat-sen University, Kaohsiung 804, Taiwan
$^2$ Aerosol Science Research Center, National Sun Yat-sen University, Kaohsiung 804, Taiwan
$^3$ Biodiversity Research Center, Academia Sinica, Nangang, Taipei 115, Taiwan
$^4$ Green Island Marine Research Station, Biodiversity Research Center, Academia Sinica, Green Island, Taitung 951, Taiwan

ABSTRACT

This study investigated the temporospatial variation, chemical composition, and source resolution of fine particles (PM$_{2.5}$) in the southeastern seas of the Taiwan Island. 24-hr PM$_{2.5}$ was sampled simultaneously at two remote sites, the Green Island (West Pacific Ocean; WPO) and the Kenting Peninsula (northern Bashi Channel; BC), in four seasons. After sampling, the chemical fingerprints of PM$_{2.5}$ were characterized and further applied to resolve the potential sources of PM$_{2.5}$ and their contribution by using a receptor model on the basis of chemical mass balance (CMB), enrichment factor (EF), and backward trajectory simulation. It showed that PM$_{2.5}$ concentrations in winter (10.8 µg m$^{-3}$) and spring (12.0 µg m$^{-3}$) (i.e., during the period of Asian Northeastern Monsoons; ANMs) were higher than those in summer (4.0 µg m$^{-3}$) and fall (6.6 µg m$^{-3}$). In terms of chemical composition of PM$_{2.5}$, secondary inorganic aerosols (SIAs = NO$_3^-$, SO$_4^{2-}$, and NH$_4^+$) (56.7–67.2%) were the dominant component of water-soluble ions (WSIs) in PM$_{2.5}$, while crustal elements (Mg, Al, Ca, Fe, and K) (44.0–61.2%) dominated the metallic contents in PM$_{2.5}$. High EF values (>10) showed that V, Mn, Ni, Cu, and Zn were potentially contributed from anthropogenic sources. Moreover, organic carbon (OC) (0.6 µg m$^{-3}$) was superior to elemental carbon (EC) (0.3 µg m$^{-3}$) in PM$_{2.5}$. The OC/EC ratios higher than 2.0 showed the potential chemical formation of secondary organic aerosols (SOAs) in the atmosphere in winter and spring. Trajectory simulation indicated that high PM$_{2.5}$ concentrations were mostly originated from North and Central China, Japan islands, and Korea Peninsula. Major sources of PM$_{2.5}$ resolved by CMB receptor modeling were ordered as: sea salts (19.9%) > fugitive dust (19.8%) > industrial boilers (oil-fired) (10.8%) > secondary sulfate (9.8%) > mobile sources (8.0%).

Keywords: Marine fine particles (PM$_{2.5}$), Temporospatial variation, Chemical composition, Source resolution, Cross-boundary transport (CBT)

1 INTRODUCTION

With the rapid development of industrialization and urbanization in the East Asia, the energy consumption and anthropogenic pollutant discharge have been increasing significantly in the past decades, unfortunately leading to poor ambient air quality of East Asian countries (Yuan et al., 2020). Particularly, during the prevailing Asian Northeast Monsoons (ANMs), cold high-pressure air masses originating from the Mongolian Plateau transport southerly to the leeward oceans (Akinwumiju et al., 2021; Filonchyk, 2022). A large amount of natural and anthropogenic particles discharged from desert, urban, and industrial areas could reduce atmospheric visibility and deteriorate the ambient air quality of leeward areas (Przybysz et al., 2020).
The impacts of particles, particularly fine particles (PM$_{2.5}$), on human health have become an increasingly serious environmental problem (Vega et al., 2021; Sakunkoo et al., 2022). Relevant medical and epidemiological researches report that long-term exposure to PM$_{2.5}$ can cause respiratory, cardiovascular, and even carcinogenic diseases. Medical evidence also show that PM$_{2.5}$ concentration is positively correlated with the inpatient and mortality rates in the heavily polluted metropolitan areas (Siregar et al., 2022; Wang et al., 2023). Both acute and chronic diseases caused by long-term PM$_{2.5}$ exposure include asthma, cough, pneumonia, bronchitis, and high blood pressure (Sánchez-Piñero et al., 2022). In addition to human health, PM$_{2.5}$ can alter the earth's radiative forcing, change the original chemical cycles, damage the ecosystem, and affect the natural resources such as drinking water, woods, and medical products (Đorđević et al., 2020; Jain et al., 2020; Mitra and Das, 2020; Wang et al., 2020; Maciejczyk et al., 2021; Bodor et al., 2022; Linh Thao et al., 2022).

In terms of chemical composition of particulate matter, water-soluble ions (WSIs) are the dominant chemical component of atmospheric particles, commonly accounting for about 20–70% of PM$_{2.5}$ (Xiao et al., 2020). Among the ions, NH$_4^+$, NO$_3^-$ and SO$_4^{2-}$ are the main components of secondary inorganic aerosols (SIAs). Particularly, NO$_x$ and SO$_x$ can react with NH$_4^+$ to form NH$_4$NO$_3$, NH$_4$HSO$_4$, and (NH$_4$)$_2$SO$_4$ (Zhang et al., 2018; Guo et al., 2017; Lang et al., 2017; Huy et al., 2020; Luo et al., 2020; Hsu et al., 2021; Guo et al., 2021). The metallic elements including Al, K, Ca, Fe, and Mg are mostly originated from the crustal materials (Amato et al., 2014; Hsu et al., 2016; Fanizza et al., 2018; Enayati Ahangar et al., 2021; Tepe and Doğan, 2021), while Fe, Ba, Zn, Cu, and Pb come mainly from the vehicular exhaust (Jeong et al., 2019; Yang et al., 2019; Bi et al., 2020; Deng et al., 2020; Lin et al., 2020; Fang et al., 2021; Niu et al., 2021; Tseng et al., 2022). The EC in the carbonaceous content mainly comes from the incomplete combustion of biomass and fossil fuels, which strongly affects the absorption of visible light and thus promotes global warming (Dinoi et al., 2017; Wu et al., 2018; Luo et al., 2021; Yan et al., 2021; Rahman et al., 2021; Singh et al., 2022). The OC/EC ratio has been used to determine the formation of secondary organic aerosols (SOAs) (Wu et al., 2020; Feng et al., 2007; Bhowmik et al., 2021). If the OC/EC ratio was greater than 2.0, it represents the highly potential formation of SOAs (Liu, 2008; Li, 2009; Dai et al., 2018).

Asian continental outflows (ACOs) originated from North China, Korea Peninsula, and Japan Islands commonly pass through the Yellow Sea (YS) and the East China Sea (ECS), and move along the Taiwan Island to the South China Sea (SCS) (Harrison, 2020; Yuan et al., 2020; Farahani et al., 2021; Hien et al., 2021). The polluted air masses carrying air pollutants emitted from natural or anthropogenic sources in the northern Asian countries could be separated into the East and West Passages while passing through the Central Mountain Range lays north-south direction in the Taiwan Island due to its high altitude of 2,000–4,000 m. For these two passages, one enters the northern South China Sea (SCS) through the Taiwan Strait (TS) (i.e., the West Passage), and the other enters the northern SCS crossing the Bashi Channel through the southeastern open seas of the Taiwan Island (i.e., the East Passage) (Wu et al., 2020; Ravindra Babu et al., 2022; Yen et al., 2022).

Previous studies have conducted the sampling of marine PM$_{2.5}$ at the islands located at the western open seas of the Taiwan Island. They found that PM$_{2.5}$ comes mainly from the northeastern Asian countries such as China, Japan, and Korea, which transports across the TS to the northern SCS. However, very few studies focused on the sampling of PM$_{2.5}$ at the eastern open seas of the Taiwan Island. Thus, it is highly requested to sample PM$_{2.5}$ and characterize their chemical fingerprints in the East Passage. Accordingly, this study aims to simultaneously sample and analyze the chemical composition of PM$_{2.5}$ at the Green Island and the Kenting Peninsula, and to clarify the temporospatial variation, chemical composition, and source resolution of PM$_{2.5}$ in the region covering the southeastern open sea areas of the Taiwan Island.

**2 METHODOLOGIES**

**2.1 Sampling Sites**

In this study, PM$_{2.5}$ samples were collected simultaneously at the two remote sites, the Green Island (GI) and the Kenting Peninsula (KP) (see Fig. S1), located at the WPO and the North Bashi Channel, respectively, which are important international waterways connecting the ECS, the SCS.
and the Philippine Sea in the WPO. The environmental description and geographical location of the two remote sites are shown in Table S1. In order to clarify PM$_{2.5}$ distribution in the region covering the southeastern open seas of the Taiwan Island, the Green Island, the largest offshore island in the eastern side of the Taiwan Island, was selected for PM$_{2.5}$ sampling, while the Cape Eluanbi Lighthouse at the was selected at the Kenting Peninsula located at the south tip of the Taiwan Island since there is no offshore island available for PM$_{2.5}$ sampling.

The sampling location at the Green Island (GI) is situated on the top floor of a 3-story building in the Marine Research Station (MRS) established and operated by the Biodiversity Research Center (BRC) of the Academia Sinica, which is about 33 km away from the east coast of the TS, approximately 9 m above the ground, and 110 m away from the nearest coastline. The sampling location at the Kenting Peninsula (KP) is situated on the top floor of 2-story building in the Cape Eluanbi Lighthouse (CEL) which is the landmark of the Kenting National Park (KNP) located at the southernmost point of the Taiwan Island. The PM$_{2.5}$ sampling location is about 5 m above the ground and 360 m away from the nearest coastline.

Both GI and KP are in the tropical monsoon climate region with warm and humid weather all year round. The number of tourists is generally higher than the population of local residents at the two remote sites where famous sightseeing and leisure locations are in the TS.

2.2 Sampling Methods

During the sampling periods, 24-hr PM$_{2.5}$ samples were collected by PM$_{2.5}$ samplers (BGI, PQ-200) at the two remote sites on September 17–23, 2020 (Fall), January 10–16, 2021 (Winter), April 12–18, 2021 (Spring), and August 11–17, 2021 (Summer). The sampling methods are described in the Supporting Information.

2.3 Chemical Analytical Methods

The chemical composition of PM$_{2.5}$ was analyzed to quantify water-soluble ions (WSIs), metallic elements (MEs), and carbonaceous contents (CCs). The analytical methods were described in the Supporting Information.

2.4 Enrichment Factor (EF)

EF is defined as the relative abundance of metallic elements to a selected reference element (Fe) to explore the correlation between PM$_{2.5}$ and crustal materials. This study used Fe as the reference element. The definition of the EF is described in the Supporting Information.

2.5 Backward Trajectory Simulation

The backward trajectories have been widely applied to describe the transport routes of air masses which could carry air pollutants to the leeward regions via suitable weather systems (Yuan et al., 2020). The principle of backward trajectory simulation is presented in the Supporting Information.

2.6 Chemical Mass Balance (CMB) Receptor Model

The source apportionment of PM$_{2.5}$ was determined by using a receptor model based on the principle of chemical mass balance (CMB) (Bie et al., 2021; Cheng et al., 2020; Cheng et al., 2021). The principle of CMB receptor model is described in the Supporting Information.

3 RESULTS AND DISCUSSION

3.1 Temporospatial Variation of PM$_{2.5}$ Concentration

The seasonal variation of PM$_{2.5}$ concentration at the two remote sites is depicted in Fig. 1. In four seasons, the concentrations of PM$_{2.5}$ at the site GI were ordered as: spring (12.7 ± 1.3 µg m$^{-3}$) > winter (10.5 ± 4.4 µg m$^{-3}$) > fall (7.5 ± 3.1 µg m$^{-3}$) > summer (4.7 ± 1.2 µg m$^{-3}$); while those at the site KP were ordered as: winter (13.1 ± 3.2 µg m$^{-3}$) > spring (11.3 ± 1.3 µg m$^{-3}$) > fall (5.7 ± 3.4 µg m$^{-3}$) > summer (3.3 ± 1.0 µg m$^{-3}$). The seasonal average PM$_{2.5}$ concentrations in summer and fall were always lower than those in winter and spring, which was mainly attributed to the fact
Fig. 1. Seasonal variation of PM$_{2.5}$ concentrations at two remote sites during the sampling periods.

that Asian continental cold-high weather system could carry air pollutants and move southwesterly with a clockwise direction in winter and spring. The blowing of polluted air masses from the northeastern Asian continent and islands to the leeward seas by ANMs resulted in the increase of PM$_{2.5}$ concentrations. Oppositely, the two remote sites were mainly influenced by the prevailing southwesterly winds in summer and fall, which brought clean marine air to the two leeward sites, resulting in much lower PM$_{2.5}$ concentrations than those in winter and spring.

In terms of the spatial distribution of PM$_{2.5}$ concentration as illustrated in Fig. 1, the concentrations of PM$_{2.5}$ were ordered as: site GI > site KP in fall, spring, and summer, while an opposite trend was observed in winter. One possible reason was that the western part of the TS was significantly affected by the cross-boundary transport (CBT) air masses blown from the northwest, which was more polluted than the eastern part of the TS in winter. Polluted air masses could arrive at the KP, the southern tip of the TS, causing the highest PM$_{2.5}$ concentrations in winter. Another possible reason was that winter was the off-season for tourism at the Green Island (site GI) where had very few local human activities due to big waves causing by strong winds, leading to relatively lower PM$_{2.5}$ concentrations at the sites GI than those at the site KP. However, such phenomenon was not observed in spring due to more human activities in the island causing by the recovery of tourist number as the temperature rose up gradually from early April to late May. The meteorological data including wind speed, prevailing wind direction, air temperature, relative humidity, and rainfall during the sampling periods at GI and KP are summarized in Table S2.

3.2 Chemical Composition of PM$_{2.5}$

The chemical composition of PM$_{2.5}$ was further analyzed to characterize their chemical fingerprints at the two remote sites. The analytical contents included WSIs, MEs, and CCs of PM$_{2.5}$. The chemical analytical results of PM$_{2.5}$ at the two remote sites are shown in Table 1 and Fig. 2.

As depicted in Fig. 2, the seasonal trends of WSI concentrations were similar to the PM$_{2.5}$ concentrations at both sites GI and KP. Overall, WSIs accounted for 44.6–46.7% of total mass of PM$_{2.5}$ (see Table 1). Five major ions including NO$_3^-$, SO$_4^{2-}$, Cl$^-$, NH$_4^+$, and Na$^+$ were the dominant species of WSIs at the two remote sites. Among them, SIAs including NO$_3^-$, SO$_4^{2-}$, and NH$_4^+$ were highly influenced by CBT, while Cl$^-$ and Na$^+$ are recognized as sea salts emitted mainly from oceanic spray (Liu et al., 2017; Guo et al., 2020; Zang et al., 2021; Dimitriou et al., 2021). Overall, SIAs accounted for 54.9 (summer) to 66.5% (spring) and 54.8 (summer) to 63.4% (spring) of WSIs at the sites GI and KP, respectively (see Table 1). The mass ratios of SIAs/WSIs at the site KP were
Table 1. The chemical composition of PM$_{2.5}$ sample at two remote sites during the sampling periods.

<table>
<thead>
<tr>
<th>Major Contents</th>
<th>Chemical Species</th>
<th>Fall</th>
<th>Winter</th>
<th>Spring</th>
<th>Summer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water Soluble Ions</td>
<td>F$^-$</td>
<td>ND</td>
<td>ND</td>
<td>0.56 ± 0.21</td>
<td>0.50 ± 0.29</td>
</tr>
<tr>
<td></td>
<td>Cl$^-$</td>
<td>0.82 ± 0.36</td>
<td>1.02 ± 0.24</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>Br$^-$</td>
<td>0.77 ± 0.09</td>
<td>0.80 ± 0.09</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>NO$_3^-$</td>
<td>0.63 ± 0.27</td>
<td>0.42 ± 0.22</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>SO$_4^{2-}$</td>
<td>0.96 ± 0.36</td>
<td>0.82 ± 0.23</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>Na$^+$</td>
<td>1.06 ± 0.50</td>
<td>0.78 ± 0.55</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>NH$_4^{+}$</td>
<td>1.49 ± 0.69</td>
<td>2.15 ± 0.40</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>K$^+$</td>
<td>0.12 ± 0.06</td>
<td>0.03 ± 0.01</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>Mg$^{2+}$</td>
<td>0.07 ± 0.03</td>
<td>0.03 ± 0.02</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td></td>
<td>Ca$^{2+}$</td>
<td>0.11 ± 0.04</td>
<td>0.06 ± 0.04</td>
<td>ND</td>
<td>ND</td>
</tr>
<tr>
<td>Metals</td>
<td>Mg</td>
<td>60.2 ± 3.7%</td>
<td>58.8 ± 2.5%</td>
<td>58.9 ± 3.0%</td>
<td>65.0 ± 2.5%</td>
</tr>
<tr>
<td></td>
<td>Al</td>
<td>0.09 ± 0.04</td>
<td>0.08 ± 0.03</td>
<td>0.12 ± 0.05</td>
<td>0.12 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>Ca</td>
<td>0.13 ± 0.06</td>
<td>0.15 ± 0.07</td>
<td>0.20 ± 0.11</td>
<td>0.20 ± 0.11</td>
</tr>
<tr>
<td></td>
<td>Ti</td>
<td>0.07 ± 0.16</td>
<td>0.19 ± 0.09</td>
<td>0.16 ± 0.06</td>
<td>0.16 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>V</td>
<td>0.05 ± 0.03</td>
<td>0.03 ± 0.03</td>
<td>0.12 ± 0.06</td>
<td>0.12 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>Mn</td>
<td>0.04 ± 0.02</td>
<td>0.03 ± 0.02</td>
<td>0.06 ± 0.03</td>
<td>0.06 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>Fe</td>
<td>0.16 ± 0.15</td>
<td>0.19 ± 0.14</td>
<td>0.20 ± 0.23</td>
<td>0.20 ± 0.23</td>
</tr>
<tr>
<td></td>
<td>Ni</td>
<td>0.09 ± 0.04</td>
<td>0.09 ± 0.03</td>
<td>0.16 ± 0.06</td>
<td>0.16 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>Zn</td>
<td>0.28 ± 0.19</td>
<td>0.24 ± 0.06</td>
<td>0.23 ± 0.06</td>
<td>0.23 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>Pb</td>
<td>0.02 ± 0.01</td>
<td>0.01 ± 0.00</td>
<td>0.02 ± 0.01</td>
<td>0.02 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>Cr</td>
<td>0.01 ± 0.01</td>
<td>0.01 ± 0.01</td>
<td>0.02 ± 0.01</td>
<td>0.02 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>Cu</td>
<td>0.13 ± 0.07</td>
<td>0.15 ± 0.02</td>
<td>0.16 ± 0.07</td>
<td>0.16 ± 0.07</td>
</tr>
<tr>
<td></td>
<td>Cd</td>
<td>0.05 ± 0.02</td>
<td>0.05 ± 0.02</td>
<td>0.06 ± 0.03</td>
<td>0.06 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>K</td>
<td>0.24 ± 0.12</td>
<td>0.17 ± 0.11</td>
<td>0.34 ± 0.16</td>
<td>0.31 ± 0.08</td>
</tr>
<tr>
<td></td>
<td>As</td>
<td>18.2 ± 1.5%</td>
<td>17.2 ± 0.6%</td>
<td>18.6 ± 1.2%</td>
<td>17.3 ± 1.0%</td>
</tr>
<tr>
<td></td>
<td>Ba</td>
<td>60.5 ± 8.9%</td>
<td>52.0 ± 5.6%</td>
<td>62.6 ± 9.6%</td>
<td>45.5 ± 2.6%</td>
</tr>
<tr>
<td></td>
<td>Sb</td>
<td>18.2 ± 1.2%</td>
<td>18.0 ± 1.0%</td>
<td>18.2 ± 1.2%</td>
<td>18.0 ± 1.0%</td>
</tr>
<tr>
<td></td>
<td>PM$_{2.5}$</td>
<td>15.7 ± 0.9%</td>
<td>14.6 ± 1.5%</td>
<td>15.7 ± 0.9%</td>
<td>14.6 ± 1.5%</td>
</tr>
<tr>
<td></td>
<td>Crustals/Metals</td>
<td>54.1 ± 6.5%</td>
<td>52.0 ± 5.6%</td>
<td>54.1 ± 6.5%</td>
<td>52.0 ± 5.6%</td>
</tr>
<tr>
<td></td>
<td>Carbon</td>
<td>10.6 ± 0.3%</td>
<td>10.3 ± 0.8%</td>
<td>11.4 ± 0.7%</td>
<td>11.1 ± 0.8%</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.79 ± 0.32</td>
<td>0.58 ± 0.36</td>
<td>1.20 ± 0.49</td>
<td>1.46 ± 0.50</td>
</tr>
<tr>
<td></td>
<td>OC</td>
<td>0.49 ± 0.20</td>
<td>0.36 ± 0.23</td>
<td>0.82 ± 0.34</td>
<td>1.00 ± 0.34</td>
</tr>
<tr>
<td></td>
<td>TC/PM$_{2.5}$</td>
<td>10.6 ± 0.3%</td>
<td>10.3 ± 0.8%</td>
<td>11.4 ± 0.7%</td>
<td>11.1 ± 0.8%</td>
</tr>
</tbody>
</table>

Unit: μg m$^{-3}$; N.D.: Non-detectable; GI: Green Island; KP: Kenting Peninsula.
higher than those at the site KP, indicating that the Green Island as an offshore island was highly affected by both oceanic spray (i.e., Mg and K) and Aeolian dust (i.e., Ca, Al, and Fe). Additionally, the concentrations of Zn in PM$_{2.5}$ at the site GI were generally higher than those at the site KP, indicating that the Green Island was more affected by traffic emissions than the Kenting Peninsula.
Moreover, at the site KP, the concentrations of Ti in PM$_{2.5}$ in winter and spring were higher than those in summer and fall. Similar seasonal trend was observed for Cu, indicating that the two remote sites were significantly affected by ANMs carrying PM$_{2.5}$ containing anthropogenic metals in winter and spring.

In terms of enrichment factors (EF) of metallic elements, the EF values of Ca, K, Mg, Fe, and Al were always lower than 10 at the two remote sites, showing that PM$_{2.5}$ were mainly emitted from crustal sources. Oppositely, the EF values of V, Mn, Ni, Zn, and Cu were higher than 10, implying that the trace metals were originated from anthropogenic sources. In addition to the contribution of crustal sources from soil dust, the two remote sites were affected by anthropogenic sources including ship emissions (V and Ni), vehicular exhausts (Mn and Zn), and industrial emissions (Cu) (Jeong et al., 2019; Yang et al., 2019; Bi et al., 2020).

As shown in Fig. 4, the concentrations of CCs also followed similar seasonal trend of PM$_{2.5}$ concentrations. In terms of carbonaceous species, organic carbon (OC) in PM$_{2.5}$ was superior to elemental carbon (EC) at the two remote sites. Particularly in winter and spring, the mass ratios of OC and EC (OC/EC ratios) were always higher than 2.0, showing that secondary organic aerosols (SOAs) were chemically formed in the atmosphere (Rastogi et al., 2014; Dai et al., 2018). These two remote sites are located at an island and along a coastline, respectively, where no large pollution sources existed nearby, thus the concentrations of EC was relatively low, while OC was mainly formed by atmospheric photochemical reactions, resulting in the increase of OC concentrations in winter and spring.

In summary, the temporospatial variation of chemical contents in PM$_{2.5}$ followed similar seasonal trend of PM$_{2.5}$ concentrations (see Fig. 5). At both remote sites, the concentrations of WSIs in PM$_{2.5}$ ranged from 1.5 to 6.0 µg m$^{-3}$, the concentrations of MEs in PM$_{2.5}$ ranged from 0.5 to 2.3 µg m$^{-3}$, while the concentrations of CCs in PM$_{2.5}$ were in the range of 0.3–2.6 µg m$^{-3}$. It showed that WSIs were the dominant species of PM$_{2.5}$, and followed by MEs and CCs at the sites GI and KP. Although the concentrations of three major contents might be different and varied with the seasons and the remote sites, the chemical composition of WSIs, MEs, and CCs in PM$_{2.5}$ were however quite similar, showing that CBT predominated both mass concentrations and chemical species of PM$_{2.5}$ at the two remote sites while compared to local emissions, especially in winter and spring.

This study further conducted a pair T-test to analyze the correlation of PM$_{2.5}$ concentration and chemical species between the sites GI and KP in four seasons (see Table S4). The correlation...
between the two remote sites in summer was relatively higher than other three seasons, mainly due to the prevailing southwestern winds. The interferences from external sources were insignificant, causing lower cross-boundary transport contributions (28.6–29.3%) and higher impacts of local emissions. In particular, K⁺, Mg²⁺, K, and Mg were highly correlated in summer, mainly because clean air masses came from the SCS, which made higher correlations between the two remote sites.

The correlation between winter and spring was relatively low, and the cross-boundary transport contribution was as high as 65%, indicating that the two remote sites were affected by the diverse and complicated emission sources in the north, resulting in high influences of LRT on the mass concentrations and chemical species of PM₂.₅, thus their correlation became lower. In fall, the chemical species of sites GI and KP had lower correlation, mainly due to significant differences in wind direction and emission sources, but K⁺ and Mg²⁺ had higher correlations (Chansuebsri et al., 2022).

### 3.3 Variation of Transport Routes

The backward trajectories of air masses arriving at the Green Island and the Kenting Peninsula were further plotted by HYSPLIT to resolve the transport routes and potential sources of PM₂.₅ in East Asia. The starting time of the backward trajectories was set at the time having the highest hourly PM₂.₅ concentration within 24 hours in each sampling day, which was applied to simulate the past 72-hr trajectories of air masses before reaching the remote site. Fig. 6 is plotted to cluster the transport routes based on PM₂.₅ concentration during the sampling periods. In this study, the concentrations of PM₂.₅ were classified into four ranges: 0–5, 5–10, 10–15, and 15–20 µg m⁻³, to cluster the transport routes of air masses with different PM₂.₅ concentration levels.

Fig. 6 shows that the highest frequency of PM₂.₅ concentrations was in the range of 10–15 µg m⁻³ at the two remote sites. In winter and spring, the transport routes of air masses moving toward the site GI mainly came from central China, north China, northeast China, and Korea. Similar to the site GI, the transport routes of air masses moving toward the site KP mainly came from central, north, and northeast China, Korea, and Japan. However, there were only a few days (January 15–16, 2021) having the PM₂.₅ concentrations within 15–20 µg m⁻³ at the site KP in winter, whereas air masses came mainly from central China. The lowest frequently of PM₂.₅ concentrations were in the range of 0–5 µg m⁻³ at both sites, whereas air masses were transported mainly from the southwest (i.e., the SCS) and the southeast (i.e., the Philippans Sea). It showed that high PM₂.₅ concentrations were mainly affected by CBT transporting from the north, resulting in an increase of PM₂.₅ concentrations in winter and spring.
Fig. 6. Backward trajectories of air masses transported toward (a) the Green Island and (b) the Kenting Peninsula during the sampling periods.
3.4 Source Resolution of PM$_{2.5}$

The CMB receptor model was applied in this study to resolve the potential sources of PM$_{2.5}$ and their contributions at the southeastern seas of the Taiwan Island. As shown in Fig. S2 and Table S5, five dominant sources of PM$_{2.5}$ at the two remote sites were ordered as: sea salts $>$ fugitive dusts $>$ industrial boilers (oil-fired) $>$ secondary sulfate $>$ mobile sources. The dominant sources respectively accounted for 0.3–2.7 µg m$^{-3}$ of PM$_{2.5}$, and their contribution percentages were in the range of 8.6–21.2%, respectively. Sea salts were the major natural sources since the two remote sites are located at an island or along a coastline both neighboring to the seawaters. High contribution of fugitive dust might be attributed to the fact that the neighborhood of the two sites are not covered by buildings or plants, which are thus easily affected by fugitive dust causing from wind erosion. Industrial boilers (i.e., oil-fired boilers) had higher contribution in fall, winter, and spring, indicating that PM$_{2.5}$ came mainly from ship emissions and industrial sources located at the north (see Fig. S5). The contribution of secondary sulfate in winter and spring was higher than that in summer and fall, resulting from the formation of SIA$s$ in the atmosphere. The contribution of mobile sources in summer and fall were higher than those in winter and spring, indicating that ambient PM$_{2.5}$ were significantly affected by local traffics especially in summer and fall.

Particularly, in order to estimate the potential contributions of CBT, the minimum PM$_{2.5}$ levels of 3.2 and 2.2 µg m$^{-3}$ at the sites GI and KP, respectively, commonly observed in summer were taken as the background PM$_{2.5}$ concentrations. The contribution of each pollution source was deducted from the contribution of each pollution source, and the difference was assumed to be the contribution of CBT. The resolved CBT contribution ratios at the two remote sites are summarized in Table S6. The contributions of CBT to PM$_{2.5}$ at the site GI accounted for 1.4–8.7 µg m$^{-3}$ with the contribution ratios ranging from 28.6–67.9%; while the contributions of CBT to PM$_{2.5}$ at the site KP accounted for 1.0–10.4 µg m$^{-3}$ with the contribution ratios ranging from 29.3–79.7%. It clearly showed that the site KP was affected by CBT much more significantly than the site GI (see Fig. 7). The contributions of CBT to PM$_{2.5}$ at the two remote sites have shown an obvious upward trend since late fall till early spring, indicating high influence of CBT blown by ANMs. Primary and secondary PM$_{2.5}$ were transported from the northeastern Asian continents to the leeward seas by ANMs.

As shown in Fig. S3, winter and spring had denser fire spots in East Asia than fall and summer. In terms of the HYSPLIT resolved backward trajectories, we found that the transport routes of air

![Fig. 7. Contribution ratios of sources for PM$_{2.5}$ transported from LRT at two remote sites during the sampling periods.](image-url)
masses were highly correlated to the regions with densely fire spots in winter and spring. It showed that the biomass burning could emit PM$_{2.5}$ mainly from Indochina Peninsula, southern China, central China, and northeastern China (Akbari et al., 2021; Amnuaylojaroen et al., 2020).

### 3.5 Spatial Distribution of PM$_{2.5}$ Concentration and their Chemical Composition at Island and Coastal Sites in East Asia

In order to explore the spatial distribution of PM$_{2.5}$ in East Asia, this study further summarized and compared the concentrations of PM$_{2.5}$ and their chemical composition at the island and coastal sites in East Asia. Marine PM$_{2.5}$ has been sampled at the East Asian Islands including the Jeju Island, the Tuoji Island, the Beihuangcheng Island, the Yangshan Island, the Matsu Islands, the Kinmen Island, the Oki Islands, the Dongsha Islands, and the Nansha Islands. In addition to East Asian islands, PM$_{2.5}$ has been sampled at several coastal sites located at the coastal cities of Fuzhou, Xiamen, and Qingdao (see Table 2 and Fig. S4). The concentrations of PM$_{2.5}$ and their chemical composition are summarized in Table 2.

As shown in Fig. S4, the geographical locations of the Tuoji Island, the Beihuangcheng Island, and Qingdao City are close to North China where the PM$_{2.5}$ concentrations were relatively high (62.0–64.0 µg m$^{-3}$), while the Yangshan Island is a seaport where the PM$_{2.5}$ concentrations were highly affected by ship emissions. During the periods of ANMs, northeastern prevailing winds could transport PM$_{2.5}$ from northern coastal metropolitan and industrial areas to the Jeju Island, the Matsu Islands, the Kinmen Island, and the Xiamen Island, and could even transport southerly to the Dongsha Islands and Nansha Islands in the SCS. It concluded that ANMs can blow PM$_{2.5}$ to the leeward island sites in the TS, the ECS, and the SCS. Combining local anthropogenic sources at the islands with the LRT of PM$_{2.5}$ to the leeward seas demonstrated a superimposition effect, resulting in significant impacts on the local and regional air qualities, which concurred with previous literature (Li, 2009; Li et al., 2013). In summary, the concentrations of ambient PM$_{2.5}$ at the northern side of East Asia were commonly higher than those at the southern side, while the concentrations of ambient PM$_{2.5}$ at the western side were commonly higher than those at the eastern side.

### 4 CONCLUSIONS

This study explored the temporospatial variation, chemical composition, and source resolution of marine PM$_{2.5}$ in the region covering the southeastern seas of the Taiwan Island. The concentrations of PM$_{2.5}$ at the two remote sites in winter and spring were always higher than those in fall and summer. Five ionic species including NO$_3^-$, SO$_4^{2-}$, NH$_4^+$, Cl$^-$, and Na$^+$ were the dominant species in

<table>
<thead>
<tr>
<th>Sampling sites</th>
<th>PM$_{2.5}$ (µg m$^{-3}$)</th>
<th>Water-soluble Ions (µg m$^{-3}$)</th>
<th>Metallic Elements (µg m$^{-3}$)</th>
<th>Carbonaceous Content (%)</th>
<th>Chemicals/PM$_{2.5}$</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kinmen Island</td>
<td>60.42 ± 33.61</td>
<td>10.58 ± 2.34</td>
<td>0.76 ± 0.62</td>
<td>11.32 ± 5.87</td>
<td>37.50 ± 8.01</td>
<td>Li et al., 2013</td>
</tr>
<tr>
<td>Jeju Island</td>
<td>34.89 ± 11.58</td>
<td>12.90 ± 1.67</td>
<td>–</td>
<td>6.48 ± 3.82</td>
<td>55.57 ± 6.14</td>
<td>Lee et al., 2015</td>
</tr>
<tr>
<td>Tuoji Island</td>
<td>62.02 ± 44.53</td>
<td>28.44 ± 22.83</td>
<td>–</td>
<td>12.21 ± 9.01</td>
<td>65.52 ± 13.32</td>
<td>Zhang et al., 2016</td>
</tr>
<tr>
<td>Beihuangcheng Island</td>
<td>63.08 ± 39.04</td>
<td>26.02 ± 20.73</td>
<td>0.81 ± 0.72</td>
<td>7.24 ± 5.20</td>
<td>53.88 ± 12.15</td>
<td>Zong et al., 2018</td>
</tr>
<tr>
<td>Yangshan Island</td>
<td>44.01 ± 28.82</td>
<td>17.83 ± 3.08</td>
<td>0.35 ± 0.04</td>
<td>1.04 ± 0.26</td>
<td>43.68 ± 1.12</td>
<td>Mamoudou et al., 2018</td>
</tr>
<tr>
<td>Fuzhou</td>
<td>30.48 ± 14.83</td>
<td>13.49 ± 1.87</td>
<td>–</td>
<td>7.01 ± 2.28</td>
<td>67.24 ± 2.83</td>
<td>Liu et al., 2020</td>
</tr>
<tr>
<td>Xiamen Island</td>
<td>32.80 ± 18.90</td>
<td>9.62 ± 1.41</td>
<td>3.83 ± 0.40</td>
<td>7.76 ± 1.84</td>
<td>87.50 ± 1.71</td>
<td>Wu et al., 2020</td>
</tr>
<tr>
<td>Matsu Islands</td>
<td>26.20 ± 2.21</td>
<td>11.02 ± 1.03</td>
<td>3.42 ± 0.34</td>
<td>2.93 ± 0.84</td>
<td>66.23 ± 3.32</td>
<td>Yuan et al., 2020</td>
</tr>
<tr>
<td>Oki Islands</td>
<td>17.93 ± 10.42</td>
<td>–</td>
<td>1.24 ± 1.11</td>
<td>–</td>
<td>6.73 ± 1.62</td>
<td>Ikemori et al., 2021</td>
</tr>
<tr>
<td>Qingdao</td>
<td>64.05 ± 39.50</td>
<td>29.30 ± 10.55</td>
<td>–</td>
<td>11.89 ± 5.81</td>
<td>64.30 ± 14.40</td>
<td>Zhang et al., 2021</td>
</tr>
<tr>
<td>Dongsha Islands</td>
<td>13.24 ± 2.52</td>
<td>4.23 ± 1.93</td>
<td>2.23 ± 0.94</td>
<td>1.42 ± 0.31</td>
<td>59.13 ± 2.31</td>
<td>Yen et al., 2022</td>
</tr>
<tr>
<td>Nansha Islands</td>
<td>5.50 ± 1.61</td>
<td>2.54 ± 0.94</td>
<td>0.90 ± 0.41</td>
<td>0.60 ± 0.23</td>
<td>72.72 ± 2.71</td>
<td>Yen et al., 2022</td>
</tr>
<tr>
<td>Green Island</td>
<td>8.86 ± 3.50</td>
<td>4.06 ± 1.21</td>
<td>1.59 ± 0.50</td>
<td>0.98 ± 0.31</td>
<td>73.80 ± 2.05</td>
<td>This study, 2022</td>
</tr>
<tr>
<td>Kenting Peninsula</td>
<td>8.34 ± 4.60</td>
<td>3.78 ± 1.06</td>
<td>1.44 ± 0.41</td>
<td>0.90 ± 0.28</td>
<td>72.20 ± 2.08</td>
<td>This study, 2022</td>
</tr>
</tbody>
</table>
the WSIs of PM$_{2.5}$, while SIAs accounted for 54.8–66.5% of WSIs. Crustal elements (Mg, K, Ca, Al, and Fe) dominated the metallic content of PM$_{2.5}$, and the C/M ratios ranged from 45.5 to 63.9%. Trace metals (V, Mn, Ni, Zn, and Cu) were originated from anthropogenic sources, including ship emissions (V and Ni), vehicular exhausts (Mn and Zn), and industrial emissions (Cu). The OC/EC ratios higher than 2.0 commonly occurred in winter and spring showed that PM$_{2.5}$ potentially contained SOAs which were chemically formed in the atmosphere due to the LRT of polluted air masses via ANMs. Highly occurred PM$_{2.5}$ concentrations were mainly in the range of 10–15 µg m$^{-3}$ and affected by CBT of polluted air masses from the northern regions to the leeward seas. The major sources of PM$_{2.5}$ at the two remote sites were ordered as: sea salts > fugitive dusts > industrial boilers (oil-fired) > secondary sulfate > mobile sources. The concentrations of CBT contributed to PM$_{2.5}$ at the site GI ranged from 1.4 to 8.7 µg m$^{-3}$, while the concentrations of CBT contributed to PM$_{2.5}$ at the site KP ranged from 1.0 to 10.4 µg m$^{-3}$. The transport routes of air masses were also correlated to the regions where the fire spots were commonly concentrated in winter and spring. Local emissions of PM$_{2.5}$ at the leeward island sites as well as the LRT of PM$_{2.5}$ from the north both have posed a typical superimposition effect.

ACKNOWLEDGEMENTS

This study was performed under the auspices of Ministry of Science and Technology (MOST) in Taiwan, Republic of China (ROC) with the research contract number of MOST109-2111-M-110-001. The authors are grateful to MOST for its constant financial support. Special thanks also go to Marine Biodiversity Research Station of Academia Sinica of ROC in the Green Island as well as the Cape Eluanbi Lighthouse at the Kenting National Park which was established and operated by Ministry of Economic Affairs (MEA) of ROC.

SUPPLEMENTARY MATERIAL

Supplementary material for this article can be found in the online version at https://doi.org/10.4209/aaqr.220350

REFERENCES

Amnuaylojaroen, T., Inkom, J., Janta, R., Surapipith, V. (2020). Long-range transport of Southeast Asian PM$_{2.5}$ pollution to northern Thailand during high biomass burning episodes. Sustainability 12, 10049. https://doi.org/10.3390/su122310049


Tepe, A.M., Doğan, G. (2021). Chemical characterization of PM2.5 and PM2.5–10 samples collected...