Spatiotemporal Variation and Chemical Fingerprints of Marine Fine Particles (PM$_{2.5}$) at the Matsu Islands in Northern Taiwan Strait

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ABSTRACT

This study characterized the PM$_{2.5}$ in terms of its spatial distribution, chemical composition, and seasonal/diurnal variation on the Matsu Islands in the northern Taiwan Strait. The PQ-200 samplers were employed to conduct simultaneous 24-h seasonal measurements of PM$_{2.5}$ on four offshore islands, viz., Nankan Island (Site NK), Beigan Island (Site BG), Dongyin Island (Site DY), and Chiukung Island (Site CK) as well as 12-h diurnal measurements for both regular and intensive samplings, and 24-h episodic measurements at one of the locations (Site NK). Additionally, the chemical signatures of PM$_{2.5}$ collected at six predominant local sources were established via chemical analysis, which coordinated as source profiles for chemical mass balance (CMB) receptor model to resolve the source apportionment of PM$_{2.5}$. Of the four seasons, summer exhibited the lowest average PM$_{2.5}$ concentration. In winter and spring, the level of PM$_{2.5}$ significantly rose under the influences of Asian Northeastern Monsoons (ANEMs). Furthermore, the spatial distribution of PM$_{2.5}$ across the islands showed a tendency to gradually decrease from the west to the east, with the Site NK always displaying the highest value regardless of the season. In general, the chemical composition primarily consisted of water-soluble ions (WSIs) owing to the abundancy of secondary inorganic aerosol (SIA) which accounted for 69.8% of the WSIs and 45.5% of the PM$_{2.5}$. Crustal elements dominated the metallic components, although the content of trace metals increased in both concentrations and proportion during the ANEMs. Organic carbon (OC) dominated the carbonaceous content of PM$_{2.5}$ in all seasons, with OC/EC values ranging from 1.64 to 3.78. During the episodes of poor air quality, the majority of incoming air masses followed a northern transport route (N-route) or an anticyclonic outflow route (AO-route) transported from the continent to the islands. The CMB receptor modeling results showed that the major sources of PM$_{2.5}$ on the Matsu Islands were road dust, industrial boilers, secondary aerosols, vehicular exhausts, and sea salts. Overall, the PM$_{2.5}$ pollution was originated from both local and remote sources, with 28~68% of the PM$_{2.5}$ concentrations arising from long-range transport.

Keywords: Matsu Islands; Marine fine particles; Spatiotemporal variation; Chemical fingerprints; Source apportionment.

INTRODUCTION

With vigorous economic development, industrialization and urbanization of East Asian countries had become more prosperous and densely populated, which lead to the increases of both energy consumption and air pollution emission (Xu et al., 2020a). For the past decades, China has experienced fast economic and industrialized development, resulting in severe environmental problems, particularly poor regional air quality. One of the most important air pollution issues concerned by general publics is regional hazes which frequently occurred in China are commonly accompanied with monsoons and could transport regionally and even globally (Kothai et al., 2008; Chang et al., 2018). Hazes are derived mainly from the formation of fine particles (PM$_{2.5}$; d$_{p}$ ≤ 2.5 µm) in the atmosphere, causing nationwide visibility attenuation (Lee et al., 2005). Moreover, hazes could not only lead to the deterioration of ambient air quality but also cause adverse effects on human health and climate change (Anderson et al., 1992; Dockery et al., 1993; Pope et al., 2002; Seinfeld and Pandis, 2006; Xu et al., 2020b).

PM$_{2.5}$ could be attributed from both natural sources and anthropogenic activities. Among them, combustion is one of the most important processes that generate fine particles. In addition to industrial combustion process, biomass burning such as slash-and-burn, and bush and forest fires could emit huge amounts of fine particles to the atmosphere (Ramanathan et al., 2007). Moreover, secondary fine particles could be also formed physically and chemically in the atmosphere, resulting in intrinsic hazards to human health and ecosystems as well (Schwartz, 1994; Seaton et al., 1995). Previous literature reported that fine particles could cause respiratory
diseases such as asthma or allergy-induced symptoms especially for children and elderly (Pope et al., 2002; Hoyt and Gerhart, 2004). Thus, fine particles have been recognized as a carcinogenic air pollutant by World Health Organization (WHO).

Long-range transport of air pollutants emitted from the upwind polluted sources in East Asia could cause the deterioration of ambient air quality in the downwind regions (Bertschi et al., 2004; Chen et al., 2014). In particular, Asian northeastern monsoons (ANEMs) constantly blow fine particles from northern China, Korea, and Japan to southern China, Japan, Okinawa, Taiwan, and even the Philippines, resulting in poor regional and ambient particulate air quality (Wang et al., 2007; Li et al., 2013a, b; Bagtasa et al., 2018, 2019; Tseng et al., 2019). Moreover, Asian dust has been proven to be transported to Taiwan Island and the Penghu Islands located in the middle of the Taiwan Strait (Yuan et al., 2004).

The Matsu Islands offshore the Minjiang Estuary face the city of Fuzhou along the coastline of southeastern China in the region of northwestern Taiwan Strait (Fig. S1). Due to its unique location, an air quality automatic monitoring station has been set up in the Dongyin Township located at the Matsu Islands by Taiwan Environmental Protection Administration (TEPA) as an alert site for monitoring upwind air pollutants moving southward. The Matsu Islands have no large-scale anthropogenic sources, which protected the islands as a background conservative environment. However, for the past decades, its ambient air quality was occasionally poor and sometimes even worse than the urban air quality of the main Taiwan Island. The Matsu Islands are situated in the northern margin of the subtropical climate monsoon weather and have an annual average temperature of 19.3°C and uneven seasonal variation of rainfall. The prevailing winds are blown from the northeast in winter and spring (from late November to early April of subsequent year), also known as Asian northeastern monsoons, while the southwestern monsoons occur mainly in summer and fall (from middle May to late September). The Matsu Islands have a population of about 12,000 inhabitants, a total area of 29.2 km². The major stationary sources are heavy-oil-fired power plants, breweries, and local gas stations. However, high PM₂.₅ concentration up to 150 μg m⁻³ has been recorded by the automatic ambient air quality station of Taiwan EPA located on Nankan Island in the winter of 2013 (TEPA, 2013). Our previous field measurement of particulate matter conducted at the Matsu Islands in the years of 2013–2014 showed that low monthly average concentrations of PM₂.₅ (< 35 μg m⁻³) were commonly observed from June to August, while high monthly average concentrations of PM₂.₅ (> 35 μg m⁻³, the 24-h PM₂.₅ standard of Taiwan) occurred from October to May of subsequent year (Chang et al., 2018). This phenomenon highly concurred with the change of prevailing winds that are recognized as East Asian monsoons, showing that prevailing wind direction is highly correlated with PM₂.₅ concentration. Poor particulate air quality (air quality index [AQI] > 100) and the prevalence of ANEMs are two major causes for high PM₂.₅ concentrations in winter and spring, which is a testimony that the impact of long-range transport of PM₂.₅ on the ambient air quality of the Matsu Islands might be underestimated and need to further study more detail, for instance its diurnal variation trend.

Previous studies showed that sulfate (SO₄²⁻), nitrate (NO₃⁻), and ammonium (NH₄⁺) are dominant ionic species of PM₂.₅ with secondary inorganic aerosol (SIA) converting from SO₂, NOₓ, and NH₃ (Brook et al., 1997; Dongarrà et al., 2007). Trace metals (such as Ba, As, Cr, Cd, Ni, and Pb) in PM₂.₅ could be emitted from the exhausts of utility power plants and waste incinerators, while crustal elements contain large amounts of Ca, Fe, Mg, K, and Al. In addition, the concentrations of trace metals including Ba, Cr, Cu, Mn, Mo, Ni, Pb, Sb, V, and Zn at the urban areas are always higher than those at the rural areas. The carbonaceous components of fine aerosols comprising elemental carbon (EC) and organic carbon (OC) both collectively are referred as total carbon (TC). Elemental carbon, also known as black carbon (BC) or graphitic carbon (GC), is chemically stable in the atmosphere and formed mainly in the combustion processes. Consequently, EC is attributed mainly to primary organic aerosol (POA), which can not only reduce the visual range due to its effective absorption of visible light, but also play an important role in the formation of secondary aerosols (Chang et al., 1982; Novakov, 1982). Elemental carbon plays as a toxic substantial media passing through the respiratory system, and thus increases the carcinogenic risk (Schwartz, 1994; Seaton et al., 1995; Ackermann-Liebrich et al., 1997). Organic carbon could be formed during the combustion process or chemical reactions in the atmosphere. Its emission sources included cooking fume, road dust, automobile exhausts, forest and bush fires, and tobacco smoking (Appel et al., 1979). Organic carbon can be divided into primary organic carbon (POC) and secondary organic carbon (SOC), which are formed by low-volatile organics from the reactions between primary carbon and volatile organics (Turpin and Huntzicker, 1995).

Accordingly, this study aims to investigate the seasonal and diurnal variation and the spatial distribution of marine PM₂.₅ concentration, and to characterize the chemical fingerprints of predominant sources at the Matsu Islands. Source identification and apportionment of atmospheric PM₂.₅ were further resolved with chemical mass balance (CMB) receptor modeling and backward trajectories.

**METHODOLOGIES**

**Sampling Protocol and Sampling Sites**

In this study, marine fine particles were simultaneously collected at four offshore island sites including Nankan Island (Site NK; 119°44ʹN, 26°10ʹE), Beigan Island (Site BG; 119°55ʹN, 26°10ʹE), Dongyin Island (Site DY; 120°29ʹN, 26°21ʹE), and Chiukung Island (Site CK; 119°93ʹN, 25°97ʹE) as shown in Fig. S1. The environmental description of these four island sites is summarized in Table 1. Both regular and intensive samplings of PM₂.₅ were conducted during the period of June 2014–May 2015. Regular sampling was conducted to periodically collect 24-h PM₂.₅ with a PQ-200 sampler at each island site in four seasons. Diurnal sampling of
PM$_{2.5}$ was solely conducted at Site NK both in the daytime (08:00–20:00) and at nighttime (20:00–08:00). Moreover, the size distribution of marine particulate matter was measured with a Micro-Orifice Uniform Deposit Impactor (MOUDI) only in winter (February 6–9) at Site NK. Intensive sampling was undertaken to collect 24-h PM$_{2.5}$ during the poor air quality periods and measure the size distribution of marine particles for continuous four days at Site NK. The PM$_{2.5}$ filters were then brought back to the Air Pollution Laboratory (i.e., the Central Laboratory) in the Institute of Environmental Engineering at National Sun Yat-sen University for further conditioning, weighing, and chemical analysis. Space drawing software (Surfer) was applied to plot monthly average PM$_{2.5}$ concentration contours for assessing the spatial distribution of marine PM$_{2.5}$ at the Matsu Islands.

Resuspension Sampling and Stack Sampling

In order to further understand the chemical fingerprints of fine particles emitted from predominant PM$_{2.5}$ sources at the Matsu Islands, this study selected six predominant sources including road dust, construction fugitive dust, stone processing, biomass burning, utility power plants, and breweries (Table S1) to collect their PM$_{2.5}$ samples. The particulate matter samples were initially sieved with Tyler 400 mesh ($d_p < 38$ µm) to collect fine dust of 5.0 g. We collected PM$_{2.5}$ samples from the six predominant sources at the Matsu Islands and brought them back to the Central Laboratory for further chemical analysis.

The dust samples obtained from road dust, construction fugitive dust, stone processing, and biomass burning were initially resuspended in a self-designed resuspension chamber (Fig. S2) to collect PM$_{2.5}$ and PM$_{2.5.10}$. The resuspension chamber with a size of 1.0 m (W) × 1.0 m (L) × 1.5 m (H) consists of a dry powder atomizer situated at the top and two dichotomous samplers stand at the bottom. After sampling, the chemical fingerprints of atmospheric fine particles, including water-soluble ions, metallic elements, and carbonaceous contents, were further analyzed (Yatkin and Bayram, 2008). Additionally, PM$_{2.5}$ samples emitted from the stacks of industrial boilers burning heavy oil and diesel for a utility power plant and a brewery were also collected. Fine particles were sampled by following a standard method of “Sampling of PM$_{2.5}$ Emitted from Stacks” (NIEA A212.108), which was mainly adopted from U.S. EPA Method 201A.

Chemical Analysis

After sampling, quartz fibrous filters were temporarily stored in an environment of 4°C and then transported back to the Central Laboratory for further conditioning, weighing, and chemical analysis. One quarter of the PM$_{2.5}$ filter was initially cut for the analysis of water-soluble ionic species. Each PM$_{2.5}$ filter was put inside a 15-mL PE bottle with distilled deionized water (D.I. H$_2$O) for ultrasonic vibration for at least 60 min. An ion chromatographer (DX-120; Dionex) was used to analyze the concentration of major anions ($\text{F}^-$, $\text{Cl}^-$, $\text{SO}_4^{2-}$, and $\text{NO}_3^-$) and cations ($\text{NH}_4^+$, $\text{K}^+$, $\text{Na}^+$, $\text{Ca}^{2+}$, and $\text{Mg}^{2+}$).

Another one quarter of the PM$_{2.5}$ filter was digested for analyzing the metallic elements of PM$_{2.5}$ by using a microwave digestion method. The PM$_{2.5}$ filter was put in a 30-mL mixed acid solution (HNO$_3$:HCl = 3:7), heated it up to 150–200°C for 2 h, and then diluted to 25 mL with D.I. H$_2$O. Fifteen metallic elements of PM$_{2.5}$ including Mg, K, Ca, Cr, Mn, Fe, Zn, Al, Cd, Pb, Ni, As, Ti, Cu, and V were analyzed with inductively coupled plasma atomic emission spectroscopy (ICP-AES; Optima 2000 DV; PerkinElmer).

Two one-eighths of the PM$_{2.5}$ filter were further used to analyze the carbonaceous content of marine PM$_{2.5}$. The carbonaceous content including elemental and total carbons were measured with an elemental analyzer (EA; EA1108; Carlo Erba). Prior to sampling, the quartz fibrous filters were preheated at 900°C for 1.5 h to expel carbon impurities from the quartz fibrous filters. The preheating procedure could minimize the background carbon in the quartz fibrous filter matrix, which might cause potential interferences with the analytical results, leading to an overestimation of the carbonaceous content of PM$_{2.5}$. The EA was operated in the procedure of oxidation at 1,020°C and followed by the procedure of reduction at 500°C, with continuous heating for 15 min. Moreover, after sampling, one-eighth of the quartz fibrous filter was heated in advance using nitrogen gas (N$_2$) at 340–345°C for at least 30 min to expel OC fraction from the filters, after which the amount of EC in PM$_{2.5}$ was determined. Another one-eighth of the quartz fibrous filter was analyzed without heating to obtain TC of PM$_{2.5}$. The amount of OC can then be estimated by subtracting EC from TC.

Quality Assurance and Quality Control

The quality assurance and quality control (QA/QC) for both PM$_{2.5}$ sampling and chemical analysis were also conducted in this study. Prior to conducting PM$_{2.5$ sampling, the volumetric flow rate of each PM$_{2.5}$ sampler was firstly calibrated with a film flowmeter (MCH-01; Sensidyne). A quartz fibrous filter was then carefully handled and placed in the PM$_{2.5}$ sampler to prevent potential cracking and dropping during the sampling procedure. After sampling, aluminum foil was used to fold the quartz fibrous filters, which were then temporarily stored at an environment of 4°C and transported back to the Central Laboratory for further chemical analysis. The sampling and analytical
procedure was similar to that described in previous studies (Cheng and Tsai, 2000; Lin, 2002; Yuan et al., 2006; Tsai et al., 2008; Tsai et al., 2010).

In this study, both field and transportation blanks were undertaken for PM$_{2.5}$ sampling, while reagent and filter blanks were applied for the chemical analysis of PM$_{2.5}$. The determination coefficient ($R^2$) of the calibration curve for each chemical species was required to be higher than 0.995. Background contamination was routinely monitored by using operational blanks (unexposed filters), that were proceeded simultaneously with the field samples. Overall, the background interference was insignificant in this study, and can thus be ignored. At least 10% of the samples were analyzed by spiking with a known amount of metallic and ionic species to determine their recovery efficiencies.

**Chemical Mass Balance Receptor Modeling**

The source apportionment of PM$_{2.5}$ was resolved by using a receptor model based on the principle of chemical mass balance (Ke et al., 2007; Kothai et al., 2008; Wang et al., 2008; Yatkin and Bayram, 2008). Since the detailed description of CMB receptor model (e.g., CMB8) is available elsewhere, only a brief summary is presented below. The CMB receptor model uses the emission profiles of prominent sources (i.e., source profile) to estimate the types of sources and their contribution to a specific receptor.

The CMB receptor model basically simulated a least-squares solution to a set of linear equations. This solution expresses each receptor’s contribution of chemical species as a linear summation of the products of source profiles and source contributions. Source profiles (the fractional amount of each species in the emissions from each source type) and receptor concentrations, each with realistic uncertainty estimates, served as input data to the CMB receptor model. The model output consists of the contribution from each source type to the total ambient aerosol mass, as well as to the concentration of individual chemical species. The CMB receptor model results are evaluated by several fit indices, such as $R^2$ ($\geq 0.8$), $p^2$ ($\leq 4.0$), $t$-statistics ($\geq 2.0$), and the percentage of mass accounted for 80–120%. The source profiles used in this study were mainly obtained from the chemical composition of PM$_{2.5}$ sampled locally at the Matsu Islands and Taiwan as well.

**Backward Trajectory Simulation**

In order to trace air masses transported towards the sampling sites at the Matsu Islands, backward trajectories were plotted by using global wind field provided by National Oceanic Atmospheric Administration (NOAA). A Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) simulation has been widely applied to simulate and plot the trajectory of a single air parcel towards a specific reception location at three heights above the ground (i.e., 100, 300, and 500 m) over a period of time. For this particular study, 120-h backward trajectories of air parcels transported towards the reception island sites in different sampling days were plotted to identify the potential source areas of marine PM$_{2.5}$.

**RESULTS AND DISCUSSION**

**Temporal Variation of Prevailing Winds**

The seasonal and diurnal variations of wind roses during the period from June 2014 to May 2015 obtained from the Matsu Meteorological Station located at Site NK are illustrated in Fig. S3. It clearly showed that the seasonal prevailing wind direction was predominated by East Asian monsoons. In summer, the prevailing winds came mainly from the southwest which blew clean air masses from South China Sea towards the Matsu Islands, while in fall, winter, and spring, the prevailing winds came mainly from the northeast where polluted air masses were blown from northern China, Japan, and Korea. Unlike the other three seasons, a daily wind direction change was observed in the season of spring (Fig. S3). The diurnal variation of wind directions in spring might not be simply caused by the sea-land breeze, but be also caused by the seasonal changes from spring to summer. One of the evidences was that the wind roses were very much similar both in the daytime and at nighttime. Another reason was that the size of the Matsu Islands might not be large enough to result in the sea-land breeze.

In addition to the seasonal variation of prevailing winds, the diurnal variation of prevailing winds at the Matsu Islands was further investigated. As illustrated in Fig. S3, there were no significant differences between the daytime and nighttime wind direction year-round at the Matsu Islands. This implied that the sea-land breeze was insignificant at the offshore islands near the Minjiang Estuary.

**Seasonal and Diurnal Variation of Marine PM$_{2.5}$ Concentrations**

In this study, both regular and intensive samplings were conducted to collect marine fine particles at four island sites in the Matsu Islands. The regular sampling of marine PM$_{2.5}$ was conducted from June 2014 to May 2015. The field-measured PM$_{2.5}$ concentrations were then used to investigate the spatial distribution and temporal variation (both seasonal and diurnal variation) of PM$_{2.5}$ concentration, and for further chemical analysis. Table 2 summarizes the seasonal and yearly average PM$_{2.5}$ concentration at four island sites at the Matsu Islands. It showed that the seasonally averaged PM$_{2.5}$ concentrations at the Matsu Islands in summer, fall, winter, and spring were $15.9 \pm 2.2$, $28.1 \pm 11.3$, $33.6 \pm 11.4$, and $26.9 \pm 9.5$ µg m$^{-3}$, respectively. In terms of different islands, a seasonal trend of winter $>$ fall $>$ spring $>$ summer was observed at Dongyin Island and Chiukung Island, while a different seasonal trend of winter $>$ spring $>$ fall $>$ summer was observed at Nankan Island and Beigan Island (Table 2). Nevertheless, the highest and lowest seasonally averaged marine PM$_{2.5}$ concentrations were consistently observed in winter and summer, respectively.

Furthermore, the seasonally averaged PM$_{2.5}$ concentration data measured at the four island sites was further applied to plot the seasonal concentration contours of PM$_{2.5}$ at the Matsu Islands. Fig. 1 depicts the seasonal variation of PM$_{2.5}$ concentration contours in the offshore island region of the Minjiang Estuary. Compared to the lowest seasonally averaged PM$_{2.5}$ concentration occurred in summer, relatively
Table 2. Seasonal and yearly average PM$_{2.5}$ concentrations at four island sites at the Matsu Islands.

| Sampling site      | Summer   | Fall     | Winter   | Spring   |
|-------------------|----------|----------|----------|----------|
| Nankan Island     | 15.6 ± 1.9 | 28.4 ± 12.5 | 34.6 ± 13.4 | 29.2 ± 9.6 |
| Beigan Island     | 16.0 ± 2.9 | 30.4 ± 12.2 | 35.8 ± 11.4 | 31.1 ± 11.1 |
| Dongyin Island    | 17.1 ± 2.1 | 26.5 ± 12.6 | 30.4 ± 11.1 | 22.4 ± 7.9 |
| Chiukung Island   | 14.7 ± 1.7 | 27.1 ± 9.5  | 33.4 ± 10.5 | 25.0 ± 5.9  |
| All islands       | 15.9 ± 2.2 | 28.1 ± 11.3 | 33.6 ± 11.4 | 26.9 ± 9.5  |

Unit: µg m$^{-3}$.

Moreover, the size distribution of marine particulate matter on February 6–9 at Site NK is illustrated in Fig. S4. It showed that a bimodal distribution of particulate matter was observed. Among four sampling days, three days showed the concentrations of fine particles were higher than those of coarse particles, while only one day (February 9) showed an opposite trend. The results indicated that PM$_{2.5}$ dominated marine particulate matter at Site NK.

Spatial Distribution of Marine PM$_{2.5}$ Concentrations

From the perspective of spatial distribution, the concentrations of marine PM$_{2.5}$ at the Matsu Islands showed a consistent descending trend from the west to the east in all seasons. While watching the spatial distribution trend more closely, we found that a spatial distribution trend of PM$_{2.5}$ concentrations decreasing from the north to the south in fall and winter, while an opposite spatial variation trend was observed from the south to the north in spring and summer. The concentrations of PM$_{2.5}$ at Dongyin Island, the farthest offshore island away from the Minjiang Estuary, were always lower than the other three islands at the Matsu Islands year-round. The spatial distribution of PM$_{2.5}$ concentration was however not well consistent with the seasonal prevailing
Fig. 2. Diurnal variation of PM$_{2.5}$ concentration during the sampling periods at the Nankan Island.

wind, showing that marine PM$_{2.5}$ transported towards the Matsu Islands was not solely influenced by long-range transport from up north, but could be also affected by local emissions from the closest metropolitan area, Fuzhou, that is located at the western range of the Matsu Islands.

Furthermore, HYSPLIT was then used to plot the backward trajectories of air masses moving towards the Matsu Islands. For this particular study, 120-h backward trajectories starting from Nankan Island at three heights of 100, 200, and 300 m above the ground were plotted to simulate the transport routes of air parcels. As depicted in Fig. S5, four typical transport routes were clustered from air masses transported towards the Matsu Islands during the PM$_{2.5}$ sampling periods. The northern transport route (N-route) was mainly transported from Siberia, Mongolia, and northern China to the eastern coastal China and arriving at the Matsu Islands (Fig. S5(a)). The southern transport route (S-route) describing air masses came mainly from South China Sea by passing through the Taiwan Strait (Fig. S5(b)). The eastern transport route (E-route) came mainly from the West Pacific Ocean by passing through the offshore of northern Taiwan Island (Fig. S5(c)). The anticyclonic outflow transport route (AO-route) transported from northern China, passed through East China Sea, and arrived at the Matsu Islands (Fig. S5(d)).

The concentrations of PM$_{2.5}$ sampled at the Matsu Islands for various clustered routes were ordered as: N-route > AO-route > S-route > E-route (Fig. 3). The results indicated that the highest PM$_{2.5}$ concentrations mostly occurred for the N-route originating from Siberia, Mongolia, northern China, and passing along the coastal region of eastern China that mainly occurred from early fall to early spring (Fig. 6(a)). On the contrary, the lowest PM$_{2.5}$ concentrations mostly occurred for the E-route originating from the West Pacific Ocean (Fig. 6(c)) since almost no anthropogenic sources in the oceans except for very low frequency of marine shipping over the West Pacific Ocean.

Previous literature reported that the level of marine PM$_{2.5}$ concentration was highly influenced by meteorological condition (Yuan et al., 2014; Li et al., 2016; Chang et al., 2018). It showed that the concentrations of marine PM$_{2.5}$ in spring, fall, and winter were much higher than those in summer. Backward trajectory simulation results indicated that the concentrations of marine PM$_{2.5}$ for the prevailing winds blown from the northeast were generally higher than those blown from the south.

Chemical Composition of Marine PM$_{2.5}$

After conducting the collecting, conditioning, and weighing of marine PM$_{2.5}$, the chemical analysis of PM$_{2.5}$ was further employed to measure the chemical composition of marine fine particles. The water-soluble ionic content accounted for 37.4–52.9% of marine PM$_{2.5}$ at the Matsu Islands. As illustrated in Fig. 4, the most abundant water-soluble ionic species of PM$_{2.5}$ were SO$_{4}^{2-}$, NO$_{3}^{-}$, and NH$_{4}^{+}$ (i.e., SIAs) at the Matsu Islands. The dominant chemical compounds in PM$_{2.5}$ were presumably ammonium sulfate ((NH$_{4}$)$_{2}$SO$_{4}$) and ammonium nitrate (NH$_{4}$NO$_{3}$) (Yao et al., 2003; Han et al., 2007; Kocak et al., 2007; Tsai et al., 2012). SIAs were not only contributed from the offshore marine emissions, but could be also emitted from inland anthropogenic sources. The mass ratios of SIAs to WSIs, i.e., SIAs/WSIs, ranged from 57.1% to 79.6% with an average of 69.8%.

The major SIAs were further used to determine the neutralization ratio (NR), sulfur oxidation ratio (SOR), and nitrogen oxidation ratio (NOR) of marine PM$_{2.5}$ as shown below (Solomon and Moyers, 1984):

$$
NR = \frac{[\text{NH}_4^+]}{[\text{nss-SO}_4^{2-}] + [\text{NO}_3^-]}
$$

where the units of [NH$_4^+$], [nss-SO$_4^{2-}$], and [NO$_3^-$] are the equivalent concentration (µeq L$^{-1}$). The NRs of marine PM$_{2.5}$ were much lower than unity in all seasons, suggesting
Fig. 3. The concentrations of PM$_{2.5}$ in air parcels moving towards the Matsu Islands for different transport routes.

Fig. 4. Seasonal variation of water-soluble ionic concentration of PM$_{2.5}$ sampled at the Matsu Islands.

that NH$_4^+$ cannot solely neutralize non-sea-salt sulfate (nss-SO$_4^{2-}$) and nitrate (NO$_3^-$). These results indicated that marine PM$_{2.5}$ was recognized as acidic fine particles at the Matsu Islands.

The units of variables S and N in Eqs. (2) and (3) are the equivalent concentration (µeq L$^{-1}$). The seasonal variation of SOR and NOR of marine PM$_{2.5}$ is illustrated in Fig. 5. As shown in Fig. 5, a significant trend for the seasonal variation of SOR in marine PM$_{2.5}$ was observed, while the seasonal variation of NOR in marine PM$_{2.5}$ was not significant year-round. We found that the seasonally averaged SORs of marine PM$_{2.5}$ were always higher than 0.25 (Colbeck and Harrison, 1984) in the seasons of winter and spring, while those were mostly lower than 0.25 in the seasons of summer and fall. This indicated that aged acidic particles could be formed in the atmosphere of the Matsu Islands in winter and spring due to long-range transport exerted by ANEMs that was blown from the upwind emission sources such as central and northern China, Japan, and Korea. In summer and fall, clean air masses mainly blown from South China Sea and West Pacific Ocean resulted in low SOR due to low sulfur...
Fig. 5. Seasonal variation of (a) SOR and (b) NOR in marine PM$_{2.5}$ at the Matsu Islands.

oxides in the atmosphere. Additionally, the seasonally averaged NORs of marine PM$_{2.5}$ were higher than 0.1 (Colbeck and Harrison, 1984) in the seasons of fall and winter (daytime only), while those were mostly lower than 0.1 in the other seasons. Overall, this study revealed that secondary sulfate was superior to secondary nitrate contributing to marine PM$_{2.5}$ at the Matsu Islands (Fig. 5).

The metallic content accounted for 7.7–19.3% of marine PM$_{2.5}$ at the Matsu Islands. Fig. 6 depicts the metallic content of marine PM$_{2.5}$ sampled at the Matsu Islands. This clearly showed that crustal elements (i.e., Al, Fe, Ca, K, and Mg) dominated the metallic content of marine PM$_{2.5}$ in all PM$_{2.5}$ samples year-round, while trace metals (i.e., Zn, Pb, Cr, Ni, As, and Cd) appeared mainly during the ANEM periods (from late fall to late spring of next year), indicating that the northeastern monsoons could transport marine PM$_{2.5}$ containing trace metals towards the Matsu Islands. Metals Fe, Al, and Ca are the dominant elements in the earth’s crustal particles emitted from road dusts, wind-blown dusts, and cement plants. Anthropogenic metallic elements could be transported towards the Matsu Islands through cross-boundary transport (CBT) via different transport routes by ANEMs. The crustal content of marine PM$_{2.5}$ can be further estimated by metallic elements using the following two models derived from previous literatures (Marcazzan et al., 2001; Chow et al., 1994).

\[
\text{Crustal Matter} = 1.89 \text{Al} + 2.14 \text{Si} + 1.40 \text{Ca} + 1.36 \text{Fe} + 1.20 \text{K} + 1.67 \text{Ti} \quad (4)
\]

\[
\text{Dust} = 1.89 \text{Al} + 1.66 \text{Mg} + 1.21 \text{K} + 1.40 \text{Ca} + 1.43 \text{Fe} + 2.14 \text{Si} \quad (5)
\]
Table 3 summarizes the crustal content of marine PM$_{2.5}$ at the Matsu Islands. It showed that the seasonally averaged crustal content of marine PM$_{2.5}$ ranged from 2.7 to 7.4 µg m$^{-3}$ and from 2.5 to 7.4 µg m$^{-3}$ for the above two predictive models, respectively. Overall, high crustal content of marine PM$_{2.5}$ was observed in winter and spring, while low crustal content of marine PM$_{2.5}$ was observed in fall and summer. The enrichment factors (EFs) determined from the metallic content of marine PM$_{2.5}$ is shown in Fig. 7. The results indicated that trace metals including Zn, Pb, Cr, Ni, As, and Cd were originated from anthropogenic sources as EF > 10, although crustal materials including Al, Fe, Ca, K, and Mg (EF < 10) dominated the metallic content of marine fine particles.

Overall, the carbonaceous content accounted for 7.8–21.0% of marine PM$_{2.5}$ at the Matsu Islands. The carbonaceous content of marine PM$_{2.5}$ sampled at the Matsu Islands is illustrated in Fig. 8(a). It clearly showed that the concentrations of organic carbon were always higher than those of elemental carbon at four island sites in four seasons. Previous literatures reported that EC has a chemical structure similar to pure graphite originates primarily from direct emissions from combustion sources. OC could be directly emitted from primary anthropogenic sources or formed chemically in the atmosphere, which is the dominant content of the secondary organic aerosol (SOA). Moreover, the mass ratios of OC to EC (i.e., OC/EC ratios) of marine PM$_{2.5}$ ranged from 1.6 to 3.8, which were mostly higher than 2.2 at the Matsu Islands, implying the highly possible formation and existence of secondary organic carbon (SOC) at the Matsu Islands. The diurnal variation of carbonaceous content in marine PM$_{2.5}$ is illustrated in Fig. 8(b). It showed that no consistent seasonal trend was observed during the sampling periods. However, the daytime carbon concentrations were higher than those at nighttime in spring and summer, while daytime carbon concentrations were lower than those at nighttime in fall and winter.

### Chemical Signatures of Predominant Local Sources at the Matsu Islands

This study selected six predominant PM$_{2.5}$ sources (i.e., road dust, construction site, stone processing, biomass burning, utility power plant, and brewery) at the Matsu Islands to establish the database for the chemical fingerprints of PM$_{2.5}$ emitted from these sources. Fig. 9 illustrates the ionic species, metallic elements, and carbonaceous content of PM$_{2.5}$ emitted from the predominant PM$_{2.5}$ sources. The fingerprints of road dust were mainly crustal elements including Al, Fe, Mg, and K. Construction fugitive dust was abundant of Al, Fe, Ca, and OC. Stone processing was abundant of Ca, Fe, Al, and SO$_4^{2-}$. Biomass burning was abundant of OC, K/K$^+$, and SO$_4^{2-}$. Utility power plant boilers burning heavy oil were abundant of SO$_4^{2-}$, EC, NH$_4^+$, and OC. Brewery boilers burning diesel was abundant of EC, SO$_4^{2-}$, NH$_4^+$, K, and OC.

Overall, chemical analytical results showed that water-soluble ionic species, metallic elements, and carbonaceous content accounted for 14.0–48.1%, 7.7–58.3%, and 11.6–31.4% of marine PM$_{2.5}$ for six local major sources. Metallic elements of Ca, Al, Fe, K, and Mg dominating the fraction of PM$_{2.5}$ showed that road dust, construction site, and stone process were mainly crustal materials. Open biomass burning and fuel boilers (heavy oil and diesel) belong to combustion processes, which had main ingredient of carbons (EC and/or OC), sulfate, potassium and ammonium.

### Source Apportionment and Cross-boundary Transport Contribution of PM$_{2.5}$

The source apportionment of marine PM$_{2.5}$ sampled at the

| Seasons | Summer | Fall | Winter | Spring |
|---------|--------|------|--------|--------|
| Crustal matter | 2.7 ± 0.5 | 4.0 ± 2.1 | 7.4 ± 2.7 | 6.9 ± 3.4 |
| Dust | 2.5 ± 0.7 | 3.9 ± 2.0 | 7.4 ± 2.7 | 7.1 ± 3.6 |

Unit: µg m$^{-3}$.

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Fig. 7. The enrichment factor (EF) values of marine PM$_{2.5}$ at the Matsu Islands.
Matsu Islands was further analyzed with a receptor model based on chemical mass balance (CMB8.0) that has been widely applied to identify the potential sources and apportion their contribution percentages (Ke et al., 2007; Kothai et al., 2008; Wang et al., 2008; Yatkin and Bayram, 2008). As illustrated in Fig. 10(a), the predominant sources contributed to marine PM$_{2.5}$ at the Matsu Islands year-round were road dust (both paved and unpaved dust), industrial boilers (both heavy oil and diesel boilers), secondary aerosols (both inorganics and organics), vehicular exhausts, and sea salts.

A significant seasonal variation of source apportionment of marine PM$_{2.5}$ was observed at the Matsu Islands. In summer and fall, clean marine air masses were transported towards the Matsu Islands mainly from South China Sea or West Pacific Ocean. The top three sources of fugitive dust, industrial boilers, and secondary aerosols contributed to marine PM$_{2.5}$ were 23.7 ± 7.1%, 22.4 ± 10.2%, and 16.2 ± 8.5%, respectively. In winter and spring, polluted air masses were transported towards the Matsu Islands mainly by ANEMs blown from northern and northeastern China, Japan, and Korea. The contributions of incinerators, industrial processes, petrochemical plants, and ironworks to marine PM$_{2.5}$ at the Matsu Islands were 6.7%, 14.4%, 14.9%, and 11.3%, respectively. Additionally, the contribution of biomass burning rose up significantly as high as 8–9% of PM$_{2.5}$, reflecting the frequent open burning of agricultural debris in the upwind source areas. It is worth noting that, during the ANEM periods, air masses originating from Siberia and Mongolia Plateau transported down to the south influencing areas due to high-pressure anticyclone system. Particularly, polluted air masses originating from northern and northeastern China passed along the coastal areas of East China through major industrial regions and moved across the Yangtze River Delta (YRD) and several industrial developing provinces in southeastern China.

This study further estimated the contribution of cross-boundary transport to marine PM$_{2.5}$ at the Matsu Islands. The background PM$_{2.5}$ concentration observed during the entire sampling periods was initially identified and applied to determine the contribution percentages of PM$_{2.5}$ to various sources in each season. The differences between the field-measured marine PM$_{2.5}$ concentration and the background PM$_{2.5}$ level was assumed as the contributions from the CBT. In this study, the lowest PM$_{2.5}$ concentrations of 13.2 µg m$^{-3}$
observed at Chiukung Island on August 29, 2015, was recognized as the background level of PM$_{2.5}$ at the Matsu Islands. Fig. 10(b) depicts the seasonal variation of contribution percentages from various sources of PM$_{2.5}$ that was transported cross-boundary towards the Matsu Islands. Secondary aerosols, including secondary organic carbon, sulfate, and nitrate, were formed in the atmosphere and transported towards the Matsu Islands increased in the seasons of winter and spring. A similar trend was also observed for the contribution of industrial processes, soil dust, and carbonaceous materials. Overall, the contribution of CBT to PM$_{2.5}$ transported towards the Matsu Islands in summer, fall, winter, and spring were 28.7%, 58.7%, 68.2%, and 57.5%, respectively.

CONCLUSIONS

The spatiotemporal distribution, physicochemical characteristics, and source apportionment of atmospheric fine particles were evaluated on the Matsu Islands. The maximum PM$_{2.5}$ concentrations were typically observed on Nankan Island, particularly during the ANEM periods, whereas the minimum concentrations were measured on Dongyin Island all year round. Chemical analysis of the PM$_{2.5}$ revealed SO$_4^{2-}$, NO$_3^{-}$, and NH$_4^{+}$ (SIA) to be the most abundant water-soluble ionic species, suggesting that fine particles on the islands mainly consist of ammonium sulfate and ammonium nitrate, which generally increased during spring, fall, and winter. Crustal elements (viz., Ca, Mg, Fe, K, and Al) accounted for the majority of the metallic content during all of the seasons, whereas trace metals (viz., Cd, As, Ni, and Cr) emerged only during the ANEM periods. Also, with OC forming in the atmosphere as a primary component of SOA, the OC/EC ratios generally exceeded 2.2. The CMB receptor modeling results identified road dust, secondary aerosol, and industrial boilers as the largest sources of the fine particles. Furthermore, remote emissions contributed 28–68% of the PM$_{2.5}$ concentrations, indicating the strong influence of cross-boundary transport.
Fig. 10. Seasonal variation of (a) the source apportionment of PM$_{2.5}$ at the Matsu Islands and (b) the contribution of various sources and cross-boundary transport.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at https://doi.org/10.4209/aaqr.2020.06.0307

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