Atmospheric PM$_{2.5}$ near an Urban-Industrial Complex during Air-pollution Episodes with Various Meteorological Conditions

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ABSTRACT

This study investigates the atmospheric fine particulate matter (PM$_{2.5}$) issue caused by the multi-effect of complicated sources, terrain, and meteorology at Southern Taiwan. Three sampling stations represent an urban, a rural, and a coastal sites near an urban-industrial complex. The atmospheric PM$_{2.5}$ were measured during a pollution episode from November to February, when the reference samples were collected in April. The sample was collected with a constant-flow sampler with the Federal Reference Method performance. After determining the PM$_{2.5}$ mass, their chemical compositions of ions, metals, and carbons were analyzed for the different properties caused by the multi-factors. The chemical mass balance (CMB) model was employed to evaluate the emission contributions. Additionally, an inverse trajectory model is used to analyze the pollutant transport and support the CMB results. The air-pollution episodes occurred within the winter to spring. The PM$_{2.5}$ were composed of 51–69% ions, 18–31% carbonaceous species, and 1.5–3.0% metals. The SO$_{4}^{2-}$, NH$_{4}^{+}$, and NO$_{3}^{-}$ contributed 92–96% of the ions. Most of the organic/elemental carbon ratios were low, suggesting more primary carbon emissions. The metal contents were minor and dominated by Fe and Zn. The CMB model indicated the PM$_{2.5}$ were dominated by 24% secondary SO$_{4}^{2-}$, 14.7% traffics, 8.3% petrochemical emissions, 6.8% soil dust, and 4.5% sintering plant emission. For non-episode days, the PM$_{2.5}$ were contributed by 34.9% traffics, 30% the secondary SO$_{4}^{2-}$, 10.3% secondary NO$_{3}^{-}$, and 6.8% soil dust. Nevertheless, the frequent sea-land breeze might lead to more powerful wind eddies and bring the primary PM$_{2.5}$ and the aerosol precursors from the emission areas. Consequently, the uncontrollable meteorological changings would lead to the pollution issues at the lowly convective area. Therefore, the averaging emissions of the PM$_{2.5}$ and precursors should be lowered; meanwhile, the rapid controls of the primary emissions are suggested when the high-level PM$_{2.5}$ are forecasted.

Keywords: PM$_{2.5}$, Monsoon, Wind eddies, Chemical composition, Source apportionment

1 INTRODUCTION

Industrialization and urbanization in Taiwan have produced a heavy environmental and public health issues. The emerging problems result from a small land area that cannot separate the industrial complexes from urban areas adequately, that is needed to mitigate the negative impacts. In particular, high ambient PM$_{2.5}$ and PM$_{10}$ concentrations in urban areas leads to the formation of smog that reduces visibility, increases traffic accidents and causes respiratory symptoms
Conversely, PM$_{2.5}$ and PM$_{10}$ can also be formed according to the way of a series of complicated processes. The Station B is the second largest station in the area: 38 km$^2$, latitude: 22.46N, longitude: 120.46E) and is near Dapeng Bay, which is designated as a national recreational area that actually attracts a larger number of tourists every year. The Station B is at Township (population: 54,000; area: 42 km$^2$, latitude: 22.55N, longitude: 120.54E) which is located 15 km towards east, from the Complexes 1 and 2 which are located towards the coast facing of the Taiwan Strait are close to the east side of Pingtung County forms a barrier that creates strong atmospheric eddies.

PM$_{2.5}$ and PM$_{10}$ accordingly (Yao et al., 2012), 2015; Gao et al., 2015; Li et al., 2016; Liu et al., 2019). Moreover, the hazardous and persistent organic contents, e.g., polycyclic aromatic hydrocarbons (PAHs) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), were observed on the PM$_{2.5}$ and led to potential health risk (Li et al., 2021; Shi et al., 2021; Yu et al., 2021). Apparently, the particles from soil and road dust re-suspension, seawater droplets, boiler and motor vehicle emissions, and some fugitive sources, e.g., night market (Amesho et al., 2021), are directly source-released. Conversely, PM$_{2.5}$ and PM$_{10}$ can also be formed according to the way of a series of complicated chemical and photolytic reactions in the geographic atmosphere from the released precursors. Moreover, PM$_{2.5}$ and PM$_{10}$ can also be transported to a location from remote emission sources (Li et al., 2015; Gao et al., 2016; Yu et al., 2019; Lv et al., 2021).

The symbolic components of PM$_{2.5}$ and PM$_{10}$ are water soluble ions (e.g., NH$_4^+$, NO$_3^-$, and SO$_4^{2-}$), metallic micro-particles, organic and elemental carbon particles (OC and EC, respectively) (Tu et al., 2022). There is an intense connection shown to reveal the existence between OC and EC in PM$_{2.5}$ and PM$_{10}$ accordingly (Yao et al., 2016; Huang et al., 2018; Liu et al., 2019). Moreover, the current PM$_{2.5}$ criteria published in the Taiwanese air quality standard (Yang et al., 2012) are 35 µg m$^{-3}$ (24-hr average) and 15 µg m$^{-3}$ (yearly average). There was an amendment proposal to have a yearly average PM$_{2.5}$ concentration brought down to 10 µg m$^{-3}$ from 15. Pingtung city is a major agricultural region in Taiwan. Three industrial complexes—an integrated steel manufacturing and two petrochemical productions are respectively at the southwest and west sides of Pingtung City. A very long survey shows that, these three complexes are suspected to be the main culprits that exacerbate heavy glitch in air pollution, which affects the areas in and around the neighboring Pingtung City, especially during a winter season with effective eddies. However, the information on their respective emission inputs and exact impacts is scarce.

This study was undertaken to analyze the compositions of air samples flowing between November and April, also the data was then used in a receptor model and was coupled with a backward trajectory model for more comprehensive source apportionment, which was utilized in the previous study (Wang et al., 2021). The resulting information will be useful for developing rational air pollutant emission control strategies.

2 MATERIALS AND METHODS

2.1 Sampling Stations Select

There were three sampling stations, named by station A, B, and C. They located at Pingtung City, urban township, coastal area, respectively (see Fig. 1). Station A was in the middle of the city (population: 200,000; area: 66 km$^2$, latitude: 22.67N, longitude: 120.49E), which is about 20 km towards the northeast of an integrated steel manufacturing complex (Complex 1) and a petrochemical production complex (Complex 2). The other petrochemical production complex (Complex 3) is located 15 km towards east, from the Complexes 1 and 2 and 15 km southeast of Pingtung City. The Station B is the second largest township in Pingtung County. Station C is classified as a rural township (population: 35,000 and area: 38 km$^2$, latitude: 22.46N, longitude: 120.46E) and is near Dapeng Bay, which is designated as a national recreational area that actually attracts a larger number of tourists every year. The Station B is at Township (population: 54,000; area: 42 km$^2$, latitude: 22.55N, longitude: 120.54E) which is located 15 km towards east, from the Complexes 1 and 2 and 15 km southeast of Pingtung City. The Station B is the second largest township in Pingtung County. Station C is classified as a rural township (population: 35,000 and area: 38 km$^2$, latitude: 22.46N, longitude: 120.46E) and is near Dapeng Bay, which is designated as a national recreational area that actually attracts a larger number of tourists every year. Complexes 1 and 2 which are located towards the coast facing of the Taiwan Strait are close to these two locations (Fig. 1). A high mountain range—Nandawu Mountain (height: 3,092 m)—at the east side of Pingtung County forms a barrier that creates strong atmospheric eddies...
counterclockwise) and frequent windy conditions when northeast wind is prevalent. Atmospheric eddies diminish when the wind direction shifts from northeast to southwest in February.

The sampling period ran from November 2018 to April 2019, with nine expected episode days selected based on weather forecasts (i.e., the weather that were conducive to bringing forth ambient PM$_{2.5}$ concentrations $> 35$ µg m$^{-3}$, 24-hr average).

2.2 Wind Backward Trajectory Analysis

The Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) is a computer model that is basically used to compute the air parcel trajectories in order to determine how far and in which direction is the parcel of air and subsequently air pollutants will travel. Wind trajectories at the sample station essentially help to analyze the pollutant source. It can be predominantly found through an archive trajectory that, if there is an actual occurrence of pollutant sources from an exact place or contributes few aspects to pollution. Three sample stations are independently operated in the HYSPLIT trajectory model. A new trajectory is eventually created every 6 hours, so that, the wind shifts thorough out the day and night can be taken into consideration. The quarter-degree archive meteorological data established by the Global Forecast System (GFS) Version 15 in June 2019 were used in the current study.

2.3 Sampling and Concentration Quantifications of Atmospheric PM$_{2.5}$

The simultaneous collection of air samples was done at three designated stations using two PQ 200 air particulate samplers (BGI, Boston, MA) at each station. A PTFE (diameter: 47 mm and pore size: 0.2 µm) filter was used in the 1st air particulate sampler to catch PM$_{2.5}$ for the measurements of both water-soluble ions and metals, whereas a Pall 7200 Tissuquartz® filter (diameter: 47 mm) was used in the 2nd air particulate sampler for carbon analyzes. The louvered size-selective inlet at the top of the air particulate sampler intercepted large particles (> 10 µm), insects, and raindrops. The air then flew through a cyclone where particles of sizes > 2.5 µm were removed by the centrifugal force. Finally, the treated air entered a filter holder where PM$_{2.5}$ was deposited onto the filter, which was sandwiched inside a filter cassette. The mass concentration and chemical composition were analyzed, including water-soluble ions, carbon components, heavy metal elements and PAHs. In order to analyze the above components, this project uses two different filter papers Teflon and quartz.
The air sampling was continued for 24 hours, and a dual diaphragm vacuum pump was actually used to dispense the air through the air particulate sampler at 16.7 ± 0.7 mL min⁻¹. The filters which were removed from the air particulate samplers in the final stage of 24-hr sampling period were carefully stored at 4°C at the sampling stations even prior shipping back to laboratory for deep analyses. Both clean and used filters were stored in the laboratory for at least 48 hours at the temperature of 20–23°C and relative humidity of 30–40% before weight measurements were begun. A Mettler ultra-micro balance (Model XPR2U) coupled with an anti-static device (Model U) was used for weight measurements. The readability and repeatability of the balance are 10⁻⁷ and 1.5 × 10⁻⁷ g, respectively.

2.4 Chemical Composition Analyses

After weight measurements are done, each PTFE filter was equally cut into two pieces using a ceramic scissor. The filter piece used for water-soluble ion analyses was immersed in 10 mL deionized water and subjected to ultrasound pulsation for approximately 90 minutes. The water was then filtered through a 0.45 μm filter and then the filtrate was collected for the measurements of the ions using a Dionex ion chromatography (Model DX-120): Ca²⁺, Cl⁻, K⁺, NH₄⁺, NO₃⁻, Na⁺, Mg²⁺, and SO₄²⁻. An Agilent 7500A inductively coupled plasma mass spectrometry (ICP-MS) was used to analyze the following metals digested: Al, As, Ba, Ca, Cr, Cu, Fe, Mg, Mn, Mo, Ni, Pb, Sd, Sr, V, and Zn. An Elementar Vario Micro Cube (Elemental Microanalysis Ltd, Devon, UK) coupled with an AS 200 auto sampler and a DP 700 integrator was used to determine the carbonaceous species. The filter sample was cut into two equal parts to have a total and elementary carbon analyses, where one is for being quantified directly to the carbon contents as total carbon (TC) level in PM2.5 and the other was preheated in a 340°C oven to eradicate all OC components and remaining carbon contents were defined as elementary carbon (EC). The organic carbon (OC) content was then calculated by subtracting EC from TC.

2.5 Analysis of Water-soluble Ion Components

There are seasonal differences for sampling sites, while SO₄²⁻, NO₃⁻, and NH₄⁺ contributed about 90% of the total water-soluble ions. I value is defined as Eq. (1) for the neutralization condition of SO₄²⁻ (Seinfeld and Pandis, 1998). When the value of \( I \geq 2 \), it implies that SO₄²⁻ has been neutralized and mainly exists as (NH₄)₂SO₄. On the other hand, if \( I < 2 \), then only part of it exists as (NH₄)₂SO₄ and free sulfate ions still exist in the atmosphere.

\[
I = \frac{NH_4^+}{SO_4^{2-}}
\]

The ratio of NH₄⁺ to NO₃⁻ and SO₄²⁻ ratio is represented by the J value defined as Eq. (2). When \( J < 1 \), then NH₄⁺ is insufficient to completely neutralize NO₃⁻ and SO₄²⁻ and therefore, they are acidic. On the other hand, if \( J > 1 \), NH₄⁺ is in surplus, and NO₃⁻ and SO₄²⁻ are basic.

\[
J = \frac{NH_4^+}{2 \cdot SO_4^{2-} + NO_3^-}
\]

2.6 Chemical Mass Balance Receptor Model

The chemical mass balance (CMB) receptor model is a tool that is commonly employed to enable both qualitative and quantitative analyses of the emission contributions for the atmospheric PM₂.₅ concentrations at sampling locations. The chemical compositions of the emission sources (i.e., emission source profiles) and the field measurement data should be collected to statistically analysis, while the sampling frequency and number are limited; otherwise, a positive matrix factorization (PMF) model should be employed (Liao et al., 2021). The CMB receptor model has been widely used to study the ambient PM₂.₅ phenomena in the U.S., Europe and Asia (Viana et al., 2008; Belis et al., 2013; Hopke, 2016).
The CMB receptor model comprises the following system of equations:

\[ C_i = \sum_{j=1}^{m} a_{ij} S_j, \quad i = 1, 2, \ldots, n \]  

(3)

where \( C_i \) is the concentration of species \( i \), \( a_{ij} \) is the fraction of species \( i \) in the emissions from source \( j \). The Eq. (3) summarized (1) will produce unique \( a_{ij} \) values, provided that \( m > n \). Performance measures of the CMB receptor model refers to the previous studies (Ke et al., 2007; Kothai et al., 2008; Wang et al., 2008; Yetkin and Bayram, 2008; Li et al., 2015).

Table S1 lists the 25 significant emission source profiles (i.e., \( n = 25 \)) that were compiled for the CMB analysis which was performed in this study. There are 22 local emission source profiles and 3 emission source profiles (petroleum productions and soil dust particles) published by the U.S. Environmental Protection Agency (U.S. EPA). The CMB analysis will not produce the unique \( a_{ij} \) values, if the nature of emission sources in a category is too similar. Therefore, an elimination process is often required to be performed until there is an output of a definite \( a_{ij} \) value when emission source selection is done, (i.e., all four performance measures are satisfied). The main elimination process will continue to be active until an emission source is identified to show which produces the largest \( a_{ij} \) value. This is the base emission source selectively opted for this assessment.

3 RESULTS AND DISCUSSION

3.1 Analysis of PM$_{2.5}$ Concentration

There were nine sampling days eventually selected keeping the forecast weather as base for the months effective from November 2018 to April 2019, i.e., ambient PM$_{2.5}$ concentrations > 35 \( \mu g \) m$^{-3}$, 24-hr average. For the daily episode, three occurred in November 2018 and one in January 2019. All non-episode days occurred in 2019 (two in February and three in April).

Fig. 2 illustrates both the episode and non-episode data on PM$_{2.5}$ concentrations and compositions in three sampling stations from left to right A, B, and C. It can be seen that the wind lines are...
cross-curled a lot because of prevailing northeast wind during the three months in winter, i.e., from November to January (see Fig. 3). Conversely, the non-episode days occurred from February to April. The wind almost came from the southwest, which would occur less wind eddies during February 20–21, when the actual wind direction shifted from northeast to southwest that essentially diminished the atmospheric eddies in the region during the three months in spring period (see Fig. 4). The PM$_{2.5}$ concentrations on an average range from 35 to 68 $\mu$g m$^{-3}$ were observed during the episode days. Obviously, the PM$_{2.5}$ concentration gradually keeps increasing as the result of strength of both northeast that basically grow in the duration of winter, especially when there is midst, as which fetching additional PM$_{2.5}$ to the actual region from a remote emission sources.

The PM$_{2.5}$ weights truly measured during the month of February 2019 were fairly low when the direction of wind shifted from northeast to southwest. The strengthening southwest wind that occurred in April 2019 coincided with the doubling PM$_{2.5}$ weights measured at three sampling stations. Therefore, the PM$_{2.5}$ weight data as illustrated in Fig. 2 seems to suggest that wind direction was important in shaping the ambient of PM$_{2.5}$ concentrations in the region between November and April.

Fig. 3. The wind trajectories from sampling site B within episode days.
The wind blowing towards the northeast gradually carries the pollutants from western industrial areas and residential areas to sampling stations. Both wind of sea and land in alternate day and night in Fig. 4, the emission done from core ports can even go inland when actually the breeze from sea blows and eddies of wind caused by breeze will have a raise in soil of downstream riverbed. Periodically there will be more frequency (see Fig. 3) when there is a breeze in alternate days, this basically creates much more powerful wind eddies. The presence of strong atmospheric eddies will also disperse PM$_{2.5}$ over a large extent of area, as indicated by the similar PM$_{2.5}$ concentration distributions observed at three sampling stations. The weight distributions of PM$_{2.5}$ components were approximately from 51% to 69% for water soluble ions, 18% to 31% for EC and OC, and 1.5% to 3.0% for solid metals. The weights of water-soluble ions measured at three sampling stations showed a strong correlation with the PM$_{2.5}$ weight. The clarity of trend was very much less for EC and OC. The weights of metals remained to be low despite significant increases in PM$_{2.5}$ weights as there was momentous progress in winter season. It was highly remarkable that, few unspecified components had been accounted as high.
as 20% of the PM$_{2.5}$ weights which measured from a range of 8% to 20% approximately. These unspecified components could also include organic matter, oxygen in metal oxides, and silicon content that were not measured in this study.

### 3.2 Wind Type Influences on PM$_{2.5}$ Concentration at Sampling Stations

Fig. 5, Fig. 10, Fig. 11, and Fig. 12 are used to describe the spatiotemporal development of the atmospheric PM$_{2.5}$ concentrations during the episodes with four different meteorological types. The contour is prepared by the Surfer$^\text{®}$ 14 using the data collected from the opened air database of Taiwan Environmental Protection Administration. The 24-h PM$_{2.5}$ samples are collected once per three days at 77 air quality monitoring stations around Taiwan.

#### 3.3 High-pressure Breeze

The high-pressure breeze occurred into the sea at an event day was recorded as first time on November 5 to 9 as show in Fig. 5. The weather pattern in the northeast monsoon has a high pressure to sea typical breeze type. The sampling stations are basically in the leeward area and the wind speed is much slow, which is very easy to accumulate the pollutants and hard to spread. The inverse trajectory simulation shows that, November 6 is noticed as short-range transmission and has domestic accumulation. The inverse trajectory simulation of the three stations in this project is shown in Fig. 5. The long trajectory of the reverse trajectory shows that, the polluted air mass mainly traveled towards southward along with the northern and central coastal cities, while the short trajectory showed that the polluted air mass was accumulated and transmitted in southern cities.

Fig. 6 illustrates the weight distributions of water-soluble ions in PM$_{2.5}$. The presence of NO$_3^-$ and SO$_4^{2-}$ aerosols could be an attribute to the traffic and combustion emissions in the region that effectively released gaseous NO$_x$ and SO$_x$ as precursors. The coldest temperatures and humidity, which scales very high during the sampling period had also favored the formation of particulate NO$_3^-$ (Liu et al., 2018). The value of $I$ ranged between 0.48 and 0.60, whereas the $J$ value ranged between 0.19–0.21 (November 5 to 9), $I < 2, J < 1$, indicating NO$_3^-$ and SO$_4^{2-}$ and an acidic atmosphere.

![Fig. 5](image-url) (a) Contours of 24-h averaged PM$_{2.5}$ concentration around Taiwan within the episodes and (b) the 24-h inverse trajectories for the atmospheric PM$_{2.5}$ at the sampling site on the episode day with sea-land breeze type of meteorology.
The carbon components in PM$_{2.5}$ are ultimately formed by the incomplete combustion sources and they mostly comprise elemental carbon (EC) or black carbon (BC), organic carbon (OC) and a trace amount of carbonate. EC is an inert, and it is directly released from the emission sources. Moreover, the presence of EC in the ambient air indicates pollution mainly caused by the motor vehicle emissions in the urban areas. According to Arimoto et al. (1996) and Dao et al. (2014), the petrol vehicle emissions would release considerable carbon particles with the OC/EC ratios ranging from about 1.0 to 4.2, the OC/EC ratios were $<2.2$, suggesting that organic carbon particles were directly emitted from the primary sources.

The solid metals are basically exiting in the atmosphere as small particles, also, consequently their ambience concentration, which is very much influenced by the air mass movements and gravitational pulls. The metals accounted only for about 1.3% to 3.0% of the PM$_{2.5}$ weights for both episode and non-episode samples and Fe and Zn were the two main components (see Fig. 8). It was surmised that, the primary sources of Fe were mostly soil dust particles, whereas motor vehicle tailpipe emissions, wear and abrasion of tires and wheel brake pads were the likely emission sources of Zn, which might also be produced from the metal forging operations.

As shown in Fig. 9, the wind field in Taiwan has changed from north wind to east wind. The analysis of the wind speed and wind direction records indicate that the northeast wind prevails at the sampling stations, due to the influence of terrain and weak wind speed. It is much easy to form eddy currents in Pingtung area, which results in the accumulation of pollutants. The main pollution sources are derived sulfates, derived nitrates and traffic sources in order. There is a small proportion of soil dust at all three stations.

### 3.4 Warm Area with Weak Northeast Monsoon

The second event day was from November 13 to 19 (see Fig. 10). The weather pattern was apparently the weak northeast monsoon warm area type. Weak northeast monsoon and High-pressure breeze weather patterns were basically downwind, while the speed of this wind was...
Fig. 7. The carbonaceous species contents in PM$_{2.5}$ during sampling periods.

Fig. 8. The metal compositions in PM$_{2.5}$ during sampling periods.
Fig. 9. PM$_{2.5}$ source contributions evaluated by the CMB model during the episode and non-episode sampling periods.

Fig. 10. (a) Contours of 24-h averaged PM$_{2.5}$ concentration around Taiwan within the episodes and (b) the 24-h inverse trajectories for the atmospheric PM$_{2.5}$ at the sampling site on the episode day with the warm area within the weak northeast monsoon type of meteorology.
less than 1 m s\(^{-1}\). The diffusion conditions were very poor and tended to the pollutant accumulation near the sampling stations. The inverse trajectory simulation shows that, there is long-distance transmission and domestic accumulation on November 14\(^{th}\) to 15\(^{th}\). The inverse trajectory simulation of the three stations in this project is shown in Fig. 10.

The water-soluble ion components on PM\(_{2.5}\) episode days are shown in Fig. 6. As Sea and land wind alternates day and night, seawater spray aerosols carried towards inland by sea breeze would accumulate SO\(_4^{2-}\) aerosols in the ambient air as well. When Ammonia, which is a specific type of gas released into the atmosphere, the reaction could be a precursor that eventually facilitated the formation of ammonium salts, such as NH\(_4\)NO\(_3\) and (NH\(_4\))\(_2\)SO\(_4\) in ambient air. The value of \(I\) ranged between 0.59 and 0.65 while the \(J\) value ranged between 0.21–0.23 (November 13 to 19), \(I < 2, J < 1\), indicating NO\(_3^-\) and SO\(_4^{2-}\) and an acidic atmosphere. The OC/EC ratios in event days were < 2.2, the organic carbon particles were directly emitted from sources, that too mostly from motor vehicles. The pollution sources are derived sulfates, derived nitrates and traffic sources. The sampling stations also have a small proportion of soil dust, the power industry and the iron and steel industry. According to this analysis results of metal components, it is speculated that the main pollution sources come from the boiler industry (heavy oil combustion, metal smelting), incinerators, coal-fired boilers and other emissions.

CMB analysis shows that the accumulated pollutants might be due to the transmission of pollution from the petrochemical industry in the upper winds because the wind speed is comparatively slow. It is speculated that the source of primary organic carbon is mostly provided by local traffic sources and waste burning. Based on the above analysis, it is speculated that the warm area weather pattern also pushes pollutants from north to south directions. Pingtung County is more heavily polluted by the upper winds, and the weak northeast monsoon pattern is more likely due to slow wind speeds, which caused poor diffusion.

### 3.5 High-pressure Backflow

The third event day occurred from January 7 to 13 (see Fig. 11). The weather pattern was a high-pressure backflow type and east winds prevailed in this weather pattern, because the southwest area was downwind and the wind speed was less than 1 m s\(^{-1}\), which caused the diffusion conditions very poor. In addition, the weak northerly wind which flows in western Taiwan mainly caused pollutants to accumulate in the southwest of Taiwan. The long trajectory of the reverse trajectory shows that the polluted air mass was mainly brought in from the southeast of the Taiwan Strait, while the short trajectory also showed that the polluted air mass which was accumulated in the air area of Pingtung County. It’s speculated that the monsoon effect was not obvious, the effect of micro-airflow by sea and land wind is significant. Post November 20, it will gradually turn into the northeast monsoon pattern and once the wind speed will rise, the quality of air flow will also be moderate.

The water-soluble ion components on PM\(_{2.5}\) event day are shown in Fig. 6. Coke oven operations at the steel manufacturing facilities could actually be a source of ammonia gas (Hein and Kaiser, 2012). It is noteworthy that, there are six types of steel manufacturing facilities across Taiwan, which can have coke ovens. Port cargo ship emission is brought inland during the day by sea wind, for the sampling stations are close to the Kaohsiung Port, one of the biggest ports in Taiwan. The value of \(I\) ranged between 0.76 and 0.81, while the \(J\) value ranged between 0.22–0.23 (January 7 to 13), \(I < 2, J < 1\), indicating NO\(_3^-\) and SO\(_4^{2-}\) and an acidic atmosphere. The OC/EC ratios in event days were < 2.2, organic carbon particles were directly emitted from adequate sources, mostly from motor vehicles. Under the high-pressure backflow weather pattern, the sampling stations are estimated to be dominated by derivative sulfate and derivative nitrate. When having a comparison between the above three weather patterns, the difference is that a higher proportion of traffic source pollution is simulated, and a higher proportion of soil dust was also simulated. According to the ratio of metals, there is no metal in a high proportion on January 10 day. It is speculated that the soil dust causes more crustal elements. This analysis and the CMB analysis can be mutually confirmed. As shown in Fig. 9, the secondary sulphate, nitrate, and petrochemical industry on January 10 is fairly over 70\% of three stations under the high-pressure backflow weather pattern. It indicates that the pollutants transferred and formed PM\(_{2.5}\) from the upwind sources, e.g., petrochemical, metallurgical industries, combustion boilers, and urban area to the
Fig. 11. (a) Contours of 24-h averaged PM$_{2.5}$ concentration around Taiwan within the episodes and (b) the 24-h inverse trajectories for the atmospheric PM$_{2.5}$ at the sampling site on the episode day with the high-pressure backflow type of meteorology.

downwind reception sites. Based on the above analysis, it is shown that, due to the prevailing easterly wind and weak northeasterly wind, the high-pressure backflow wind, which causes the weather patterns A and B stations to be polluted from winds flowing upward, while the southernmost C station is mostly polluted by local pollutants. Poor diffusion and accumulation results in bad air quality.

3.6 Northeast Monsoon

On the fourth event day, the northeast monsoon pattern was from April 12 to April 16. This weather pattern was mostly northeasterly winds (see Fig. 12). At 1–2 m s$^{-1}$, it is easy to transfer the pollutants from the upper wind to the Pingtung area, resulting in the deterioration of air quality. According to the reverse trajectory simulation, the pollutants mostly get entered to Pingtung County from the north on April 14 and the northeast monsoon in western Taiwan basically causes many pollutants to eventually accumulate its source in southwest of Taiwan, which is the inverse trajectory simulation of stations as shown in Fig. 12. The long trajectory of the reverse trajectory shows that, the polluted air mass was mainly brought in from the southeast direction of the Taiwan Strait, while the short trajectory also showed that, the polluted air mass accumulated in the air area of Pingtung County. The influence of the micro-airflow effect on the sea and land peaks. After April 16, when the wind speed rises, the air quality will return to normal stage.

The water-soluble ion components on PM$_{2.5}$ event day are shown in Fig. 6. The value of $I$ ranged between 0.42 and 0.55 while the $J$ value ranged between 0.20–0.21 (April 12–16), $I < 2$, $J < 1$, indicating NO$_3^-$ and SO$_4^{2-}$ and an acidic atmosphere. The OC/EC ratios in event days were $< 2.2$, the organic carbon particles were mostly emitted from the primary sources, e.g., traffic sources, open burning, combustion boiler. Under the weather pattern of northeast monsoon type, the sampling stations are being estimated to be equally dominated by derivative sulfates and traffic sources. According to the ratio of metal, there is also a high ratio of man-made pollution on April 14. It is speculated that the main pollution sources come from the boiler industry and metal smelting, incinerators, coal-fired boilers and other emissions.

According to the CMB simulation estimation, as shown in Fig. 9, under the weather pattern of the northeast monsoon type, the three different stations are profoundly estimated to be
Fig. 12. (a) Contours of 24-h averaged PM$_{2.5}$ concentration around Taiwan within the episodes and (b) the 24-h inverse trajectories for the atmospheric PM$_{2.5}$ at the sampling site on the episode day with the northeast monsoon type of meteorology.

dominated by derivative sulfates and traffic sources. It is eventually speculated that, the keen pollution sources basically come from boiler combustion and metal smelting, incinerators, coal-fired boilers and other different emissions.

4 CONCLUSIONS

The air pollution of PM$_{2.5}$ in Pingtung County in Taiwan were investigated for its multi-conditions including meteorological, emission, and terrain factors for the episode and normal cases during November to the next April. The study reported was undertaken to mainly examine the regional PM$_{2.5}$ problems in and around Pingtung City and its surrounding communities to effectively produce the useful information for the development of rational ambient PM$_{2.5}$ control strategies. The following conclusions can be drawn.

1. The PM$_{2.5}$ composition were 51–69% of water-soluble ions, 18–31% of carbonaceous species, and 1.5–3.0% of metals. SO$_4^{2-}$, NH$_4^+$, and NO$_3^-$ were the water-soluble ions, 92 to 96% for episode samples and 75 to 95% for non-episode samples.

2. Most of the episode and non-episode samples taken at three sampling stations had their OC/EC ratios less than 2.2, suggesting that organic carbon particles were directly emitted from the primary sources, e.g., traffic sources, open burning, combustion boiler.

3. Metals were a minor component in PM$_{2.5}$. Fe and Zn were noted as the principal components also including the former associated with the soil dust particles, the latter from the motor vehicle tailpipe emissions and wear and abrasion of tires and brake pads.

4. During the episode days, the CMB results shows the atmospheric PM$_{2.5}$ were contributed by 24% of secondary SO$_4^{2-}$, 14.7% of traffics, 8.3% of petrochemical emissions, 6.8% of soil dust, and 4.5% of sintering plant emission. A maximum petrochemical-based emission was observed (13.7%) at station A during the episode days, suggesting the northeast wind would carry the PM$_{2.5}$ to the actual region. For non-episode days, the PM$_{2.5}$ were contributed by 34.9% of traffics, 30% of the secondary SO$_4^{2-}$, 10.3% of secondary NO$_3^-$, and 6.8% of soil dust.

5. The fuels used in motor vehicles and industries from the industrial and urbanization areas were equally responsible for the emission contribution observed by promoting the photolytic
formation of both NO$_3^-$ and SO$_4^{2-}$ in the atmosphere. The soil dust particles that were re-suspended from the exposed ground surfaces played a vital role in influencing the regional PM$_{2.5}$ phenomena, just because of the wind eddies caused by breeze flowing from sea land.

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**SUPPLEMENTARY MATERIAL**

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**REFERENCES**


