1 Markers of East Asian dust storms in March 2010

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Abstract

The variations in particulate matter (PM) and in eighteen metallic elements in four different particle sizes in the air of southern Taiwan were investigated from February to March 2010. The variation in mean mass concentrations of $PM_{2.5}$, $PM_{2.5-10}$, PM_{10} , and TSP between Asian dust storm (ADS) and non-dust storm (NDS) periods suggested that the dominant fractions were $PM_{2.5-10}$ and $PM_{2.5}$, respectively. The obvious differences in metallic element concentrations between ADS and NDS periods suggested that crustal elements (Fe, Mg, Sr, Co, Ba and Mn) can be used as dust storm indicators, in addition to Ca and Al. Both mass fractions and metallic concentration ratios indicated that the $PM_{2.5-10}$ fraction can distinguish between ADS and NDS periods. An enrichment factor (EF_c) demonstrated that most anthropogenic elements were significantly lower in ADS periods than in NDS periods. The EF_c values for anthropogenic elements in coarse particles may be used as indicators of dust storm invasion as well. An association was found between the extent of dust storm effects on receptors and the residence time (duration) of the event, in addition to the transport pathway.

Keywords: Particulate matter; Metallic elements; Taiwan; Indicators; Enrichment factor

1. Introduction

Particles are ubiquitous in the atmospheric environment. Under specific meteorological conditions, large amounts of material can be transported in the form of dust storms. Most Asian dust storms originate in the Gobi Desert of Mongolia, northern China, and the Taklimakan Desert (Sun et al., 2005; Zhang et al., 2010; Wang et al., 2011), and then move under the influence of the prevailing westerlies to Korea, Japan, and the west coast of North America (Kanayama et al., 2002; Kim et al., 2003; Ma et al., 2004; Zhao et al., 2008), and southward to Taiwan (Chen et al., 2004a; Liu et al., 2009) and Hong Kong (Lee et al., 2012), even reaching the South China Sea (Chuang et al., 2013). The effect of dust storms on air pollutant compositions and concentrations in downwind area depends on their sources, transport pathways, and strength (Sun et al., 2005; Sun et al., 2006; Zhang et al., 2010). During transport, anthropogenic pollutants (e.g., coal burning, vehicle emissions) that accompany aerosols from dust storms can cause the deterioration of air quality of downwind regions (Wang et al., 2007; Zhao et al., 2007; Huang et al., 2010; Liang et al., 2013). Some studies indicate that ADS are accompanied by an increased risk of respiratory and circulatory disease and pneumonia or associated cardiopulmonary diseases and non-accidental and cardiovascular deaths in Taipei (Chen et al., 2004b; Chan et al., 2008; Chan and Ng, 2011). Hong et al. (2010) also pointed out that metals adhering to ADS particles decrease pulmonary function in children during dust storm periods. In addition, dust storm aerosols also play an important role in the global climate change by affecting the radiation budget (Sokolik and Toon, 1996), the primary biological productivity in the ocean (Bishop et al., 2002), and atmospheric chemistry (Seinfeld et al., 2004). These effects on human health and environment make dust storms an

important global issue for East Asia.

Taiwan is located downwind of the dust storms arising in the East Asian continent, making it a good site for observations of the variations in particulate matter (PM) and contaminants along the transport pathways. During March 2010, two dust storm events occurred in East Asia: one was a severe dust storm from March 19th to March 23rd, generated in the Gobi Desert (Li et al., 2011). Many investigations were reported (e.g., Bian et al., 2011; Li et al., 2011; Lee et al., 2012; Tatarov et al., 2012; Chuang et al., 2013) regarding particulate matter concentration, comparisons of different monitoring instruments, and the relationship between dust storms and atmospheric circulation and model simulations. The present study seeks to improve the understanding of the evolution of different particle fractions, chemical characteristics, and indicators during non dust storm (NDS) and dust storm (ADS) periods.

The strategy used in this study was to examine eighteen metallic elements in PM_{2.5-10}, PM_{2.5}, PM₁₀ and TSP in southern coastal area of Taiwan during 2010. The temporal and spatial variations of PM and metallic elements were investigated to identify dust storm events and to map the differences between upstream and downstream characteristics in the dust storm period. The particle mass, fraction mass ratio, elemental concentrations, the ratio of PM_{2.5-10} and PM_{2.5} in PM₁₀ and enrichment factors (EF_c) were used for elucidation of the difference in chemical characteristics and distribution between NDS and ADS periods, as well as for development of indicators for identifying dust storm events. The PM₁₀ data obtained from other monitoring stations were also used to show the effects of a severe dust storm across the wider East Asian region.

2. Materials and methods

2.1. Sampling site

The sampling site (22°37'N, 120°18'E) is located at the campus of National Sun Yat-sen University (NSYSU) in Kaohsiung, which is adjacent to the coast (Fig. 1). Kaohsiung is an industrial city with a population of 2.8 million. To the north and east the site is surrounded by Shoushan Mountain. Kaohsiung Harbor is approximately 1.2 km south of the sampling site. Sampling was carried out from the rooftop of a building, approximately 25 m above the ground level and 100 m away from the shoreline of Taiwan Strait. Two power plants are located north (28 km away) and south (13 km away) of the sampling site; an industrial park is located at the southeast side (13 km away).

2.2. Sampling and analytical procedures

EPA method 1669 (USEPA, 1996) was adopted for sampling and analysis, thereby reducing contamination from sampling, storage, and analysis processes (Windom et al., 1991). All labware was soaked in 10% HNO₃ solution, rinsed with deionized water (18MΩ), and dried in a clean bench. All labware was stored in plastic bags prior to use.

A Universal Air Sampler (Model 310, MSP Corporation, USA) was used for sampling PM_{2.5} (fine) and PM_{2.5-10} (coarse) samples. The flow rate was set at 300 L min⁻¹. A high volume air sampler (PS1, TE-1000 PUF, Tisch environmental corporation, USA) was used for sampling total suspended particles (TSP). An orifice calibrator was used to calibrate the PS1 before sampling. The sampling flow rate was set at 225 L min⁻¹. Particle samples (PM_{2.5-10}, PM_{2.5}, and TSP) were concurrently collected from February to March in 2010. During the sampling period, two dust storm events were observed (3 samples). One appeared on March 16th, 2010 and the

other one was on March 21st to 22nd, 2010; the latter was the strongest dust storm ever recorded in Taiwan (<http://dust.epa.gov.tw/dust/zh-tw/Database.aspx>). The rest of samples were collected in NDS periods. A total of thirteen PM_{2.5}, PM_{2.5-10}, PM₁₀ (PM_{2.5} + PM_{2.5-10}) and TSP samples were collected for 24 h of each sampling period on quartz fiber filters (Pall, 2500QAT-UP). After sampling, filters were placed on a pre-cleaned plastic petri dishes, sealed in plastic bags, and put into a clean plastic box. The filters were first conditioned for 48 h in an electric desiccator at humidity 45±5% and temperature 25±3°C prior to weighing.

Filter samples were cut with ceramic scissors and extracted with a mixture of 8 mL nitric acid (Merck, Tracepure 69%) and 2 mL hydrogen fluoride (Merck, Proanalysis 40 %) (Chester et al., 1999; Gao et al., 2002) in a Teflon screw-cap vial (Savillex Corp., UK). Complete dissolution of samples was achieved after a period of 3–4 h digestion on a hotblock (SC 154, Environmental Express Corp., SC) at 160°C. The digested solution was then evaporated to near dryness in a clean hood and was diluted to 30 mL with 1 % nitric acid for analysis. Aluminum, Ca, Fe, Mg and Na were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES, Thermo IRIS Intrepid II XSP) and V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Sr, Cd, Ba and Pb by inductively coupled plasma mass spectrometry (ICP-MS, HP4500).

2.3. *Quality control and quality assurance*

Data quality was assured with method blanks, quality control (QC), duplicate analysis (DA), and matrix spiked samples. All the control samples were treated in the same way as the actual samples. In addition, method detection limits (MDLs) were determined as the concentration equivalent of three times the standard deviation of seven replicate measurements of the analyte in reagent water, and then converted to

atmospheric concentration in ng m^{-3} . In this study, all data reported were corrected using the method blank. The results of QC, DA, matrix spike, and MDLs were from 94.6% (Mg) to 106.6% (Ca), 1.6% (Fe) to 23.3% (As), from 91.2% (Sr) to 113.3% (Pb) and from 0.0028 ng m^{-3} (As) to 1.9 ng m^{-3} (Ca), respectively.

3. Results and discussion

3.1 Identification of dust storm events

Two dust storm events were announced by the Taiwan EPA (TEPA) during the study period, one occurred on 16th March and the other on 21st to 22nd March, 2010. Apart from the announcements from TEPA, dust storm events in the study area were identified by the analysis results of this study, such as temporal variation of PM_{10} concentrations, particle mass and size fraction ratios, aluminum element in PM_{10} (given in supplemental materials, Fig. S1), and the dust storm transport pathway established from back trajectory analysis.

3.1.1 Temporal variations of PM_{10} concentrations

Figs. 2a and 2b present the temporal variations of PM_{10} concentrations from the East Asian continent during a severe dust storm period (19th to 23rd March 2010). The back trajectories show that the dust storm originated in the Gobi Desert, moved easterly across China and the Korean peninsula (Li et al., 2011; Kwak et al., 2012) and Japan (Nagasaki), and then southerly down to South China (Shanghai and Xiamen), Hong Kong, and Taiwan (Hsu et al., 2013). The dust storm reached Gwangju (Korea) at 12:00 local time (LT) on 20th March with the highest hourly PM_{10} concentration of $1864 \mu\text{g m}^{-3}$ (Park and Cho, 2013) and Nagasaki (Japan) at 21:00 (LT) on 20th March with the highest hourly PM_{10} of $727 \mu\text{g m}^{-3}$

(<http://gissv02.pref.nagasaki.jp/TaikiWeb/MainController>). For the southerly route, the dust storm arrived at Beijing (Li et al., 2011) and Shanghai (Wang et al., 2013) on 20th March, and arrived at Xiamen (Zhao et al., 2011), Hong Kong, and Taiwan (Kaohsiung) on 21st March. The maximum hourly concentrations of PM₁₀ were 5080 $\mu\text{g m}^{-3}$, 727 $\mu\text{g m}^{-3}$, 1700 $\mu\text{g m}^{-3}$, 990 $\mu\text{g m}^{-3}$, 775 $\mu\text{g m}^{-3}$, and 791 $\mu\text{g m}^{-3}$ for Beijing, Shanghai, Xiamen, Hong Kong, and Kaohsiung, respectively. The maximum hourly concentrations of PM₁₀ revealed that concentrations increased more significantly when a city was close to the dust storm track.

Evidence from hourly PM₁₀ concentrations measured by the Taiwan monitoring stations (<http://taqm.epa.gov.tw/taqm/tw/HourlyData.aspx>) indicates that the dust storm arrived in northern Taiwan (Taipei) at 02:00 (LT) on 21st March, moved to central Taiwan (Taichung) at 08:00 (LT), and then entered southern Taiwan (Kaohsiung) at 14:00 (LT). The dust storm reached east Taiwan (Hualien) at 06:00 (LT), Taitung at 09:00 (LT), and arrived down at Pingtung (Hengchun, the southern tip of Taiwan) at 12:00 (LT). The arrival time of the dust storm and the variation in the daily mean PM₁₀ concentrations suggest that western and eastern Taiwan were affected by two different air parcels during this dust storm event in Taiwan (a detailed discussion is given in supplemental materials). In the case of Dongsha station, the back trajectory suggested that dust from southern China was the source of PM₁₀ observed at 19:00 (LT) 21st March 2010. The monitoring record from the stations show that decreasing daily mean PM₁₀ concentrations of dust are related to increasing travel times. This could be due to scavenging effects (e.g., pollutant dispersion in the atmosphere, dry and wet deposition of particles) along the transport pathway (Kim et al., 2007). The temporal variation in PM₁₀ concentration and back trajectories suggest that the dust storm observed in the study area originated from northern and

northwestern China (the Gobi Desert).

3.1.2 Particle mass concentrations and size fraction ratios

Table 1 shows the particle masses of $PM_{2.5}$, $PM_{2.5-10}$, PM_{10} , and TSP for both NDS and ADS periods. The average particle masses of $PM_{2.5}$, $PM_{2.5-10}$, PM_{10} , and TSP in NDS period were 46.0 ± 19.1 , 18.7 ± 8.7 , 64.7 ± 24.6 , and $101 \pm 45 \mu g m^{-3}$, respectively. The average particle masses of $PM_{2.5}$, $PM_{2.5-10}$, PM_{10} , and TSP in ADS periods were 60.3 ± 29.1 , 135 ± 101 , 195 ± 128 , and $358 \pm 236 \mu g m^{-3}$, respectively. These results demonstrated all the PM masses in the ADS periods were higher than those in the NDS periods. The high concentrations of $PM_{2.5}$, PM_{10} , and TSP in the ADS periods exceeded the current Taiwanese 24-hr air quality standard of 35, 125, and $250 \mu g m^{-3}$, respectively (<http://ivy5.epa.gov.tw/epalaw/docfile/040060.pdf>). In addition, particle mass ratios of ADS/NDS for $PM_{2.5}$, $PM_{2.5-10}$, PM_{10} , and TSP were 1.3, 7.2, 3.0, and 3.5 times, respectively. The $PM_{2.5-10}$ showed the highest enhancement (ADS/NDS) of particle mass among the four fractions. Chen et al. (2004a) pointed to a uni-modal distribution and peak particle sizes between 3.2 and $5.6 \mu m$ in diameter in ADS periods. Particle mass ratios in this study were similar to those reported by Chen et al., (2004a), with $PM_{2.5-10}$ as the major fraction during ADS periods.

The difference in the dominant particle size between NDS and ADS periods is shown in Table 1. In NDS periods, all the $PM_{2.5}$ masses were higher than $PM_{2.5-10}$, but the opposite situation was observed in ADS periods (the $PM_{2.5-10}$ masses were over $PM_{2.5}$). The ratios of $PM_{2.5-10}/PM_{10}$ in NDS were from 0.22 to 0.48, which were smaller than those ratios (from 0.55 to 0.73) in ADS periods (Table 1). The ratios of $PM_{2.5-10}/PM_{10}$ (in ADS) in this study were similar to those reported by Chen et al. (2004a). These results indicate that $PM_{2.5}$ and $PM_{2.5-10}$ were the dominant fractions in

PM₁₀ during NDS and ADS periods, respectively. These two fractions in PM₁₀ can therefore be used to distinguish between NDS and ADS periods in the study area.

3.2 Concentration of metallic elements

Table 2 lists the concentrations of eighteen metallic elements in PM_{2.5}, PM_{2.5-10}, PM₁₀, and TSP for NDS and ADS periods. Higher concentrations of most metallic elements are observed in ADS periods compared with NDS periods and of these, Al, Ca Fe, Mg, and Na are the most abundant, accounting for over 90% of the total. The average percentage of Na in TSP mass was 4.44% and 3.57% in NDS and ADS period, respectively. It is noted that both were higher than the average concentration of Na, 2.36%, in the continental crust (Wedepohl, 1995). This result indicates that crustal and marine aerosols were major elemental sources for the particles (Park et al., 2004; Hsu et al., 2004). During NDS periods, Zn and Pb were the two dominant trace elements in PM_{2.5}, PM_{2.5-10}, PM₁₀, and TSP, while Zn and Mn dominated during ADS periods. The concentrations of Co and Cd were the two lowest during both NDS and ADS periods. The concentrations of V and Ni in PM_{2.5} were lower in ADS than in NDS periods. However, the opposite situation was observed for PM_{2.5-10}, PM₁₀, and TSP. These results indicate that V and Ni in PM_{2.5} largely originate from local emissions, while dust storms contribute these two elements to PM_{2.5-10}, PM₁₀, and TSP.

Crustal elements (Ca, Al, Fe, Mg, Sr, Co, Ba, and Mn) display higher ADS/NDS concentration ratios when compared to ratios for anthropogenic elements (Ni, Cu, Zn, As, Se, Cd Pb, V, and Cr) for all four particles size fractions (Fig. 3). The ADS/NDS concentration ratios of crustal elements were higher in PM_{2.5-10} than in PM_{2.5}, PM₁₀ and TSP. Calcium showed the highest ratios, compared with other elements in PM_{2.5}, PM_{2.5-10}, and PM₁₀; while Al revealed the highest ratio in TSP. The high ADS/NDS

ratios for Ca found in this study was similar to the findings of Sun et al. (2005) and Kang et al. (2009). This suggests that Ca could also be used as an indicator element to distinguish NDS and ADS events in the study area. In addition, dust storm source regions and transport pathways could cause the difference of chemical composition (Ca, crustal, and anthropogenic elements) in particles (Son and Park, 2015; Wang et al., 2008; Cao et al., 2005). Dolomite was used as a tracer of Asian dust from the regions on north margin of Tibetan Plateau (Li et al., 2007). Calcium concentrations obtained from south Mongolia and north China (N) were found relatively higher than those from northeastern China (NE) in this study (Table S1 and Fig. S2). This observation was similar to that of Son and Park (2015).

The ADS/NDS concentration ratios of anthropogenic elements (Ni, Cu, As, Zn, Pb, Cd, Se, V, and Cr) are given in Fig. 3. The lowest ratio in $PM_{2.5}$ was for Ni (0.92) and the highest was Pb (1.9). For $PM_{2.5-10}$ the lowest and highest ratios were for Zn (0.77) and As (4.4), respectively. For PM_{10} and TSP, the lowest and highest ratios were both for Cu (1.1 and 0.73) and As (2.1 and 2.8), respectively. These ratios suggest that dust storms not only transport crustal and marine elements, but they also carry significant quantities of anthropogenic elements (Huang et al., 2010). Accordingly, the use of Al and Ca as indicator elements could be supplemented by measurements of the simultaneous high variations of Fe, Mg, Sr, Co, Ba, and Mn (Fig. 3).

3.3 Enrichment Factors (EF_c)

Enrichment factors (EF_c) are used to distinguish between elements in aerosols originating from natural (crustal and marine) and anthropogenic sources. Elemental EF_c values above 10 typically indicate an anthropogenic origin (Chester et al., 2000; Hsu et al., 2004). The EF_c value of element X is calculated from the equation:

$$EF_c = (C_X/C_{Al})_a / (C_X/C_{Al})_c$$

where C_X and C_{Al} are the concentrations of element X and the reference element (Al), respectively. The subscript 'a' refers to aerosol in the atmosphere, whereas 'c' refers to the crust. The average concentrations of elements in the crust were obtained from Wedepohl (1995). The EF_c values of the seventeen elements in $PM_{2.5}$, $PM_{2.5-10}$, PM_{10} , and TSP during NDS and ADS periods are shown in Figs. 4a-d. During NDS periods, the EF_c ranges from below one to thousands. The elements Cu, Zn, As, Se, Cd, and Pb were highly enriched ($EF_c > 100$) in NDS periods. This result indicates a substantial anthropogenic source for these elements in the study area.

The EF_c values of V, Ni, Cu, As, Zn, Pb, Cd, and Se in all fractions were over 10, except V ($EF_c = 5.6$) in $PM_{2.5-10}$. Therefore, these elements appear to be anthropogenic in origin. The rest of the elements ($EF_c < 10$) are likely to be crustal in origin. In ADS periods, the EF_c values are significantly lower than in NDS periods, except for Ca in $PM_{2.5}$, $PM_{2.5-10}$, and PM_{10} and for Fe and Mg in $PM_{2.5}$. This suggests that crustal elements make a substantial contribution to PM, which results in low EF_c values during ADS periods (Sun et al., 2005; Huang et al., 2010). However, Zn, As, Se, Cd, and Pb in $PM_{2.5}$ showed the highest EF_c values among all fractions during ADS periods. The sources for these elements are likely to be vehicle emission (Zn, Cd, and Pb) and coal burning (As and Se), locally emitted or adhering to ADS particles along transportation pathway (Fung and Wong, 1995; Wu et al., 2007; Lin et al., 2005; Hu et al., 2013).

Figure 4 also displays the statistical results of t-tests of the difference between NDS and ADS periods. Of the 17 elements determined, nine elements in $PM_{2.5}$, 13 elements in $PM_{2.5-10}$, 12 elements in PM_{10} , and 16 elements in TSP showed a significant difference. More specifically, Ba, Na, Cr, V, Ni, Cu, As, Zn, Pb, Cd, and Se

simultaneously showed a significant difference in $PM_{2.5-10}$, PM_{10} , and TSP. All anthropogenic elements revealed a significant difference in the coarse fractions ($PM_{2.5-10}$, PM_{10} , and TSP). This result arose because coarse particles are form the dominant fraction in ADS (Chen et al., 2004a; Tsai et al., 2012). The low EF_c during the ADS period may be attributed to the substantial presence of crustal Al accompanying ADS. This finding supports the use of the EF_c of anthropogenic elements as an indicators of dust storm incursions.

3.4 Spatial variations during a severe dust storm event

The spatial variation of PM_{10} concentrations from East Asian region to Taiwan during the severe dust storm (19th to 23rd March 2010) are shown in Fig. 2a and 2b. Two transport pathways of dust storm can be categorized. Daily PM_{10} concentrations observed at air monitoring stations in the East Asian region varied along the dust storm path (Fig. 2a). There was an obviously decrease in PM_{10} concentration as the dust storm moved to Gwangju, Korea and Nagasaki, Japan (Fig. 2a). The low daily PM_{10} concentration observed could be due to the dust storm mixing with clean maritime air, local air masses with background concentrations, and short residence times. A comparison of dust storm age among all monitoring stations suggests that the dust storm remained in Xiamen and Hong Kong (about 2 days) longer than it resided in Beijing, Shanghai, Gwangju, and Nagasaki (one day or less). Longer residence time of the dust storm accompanying dispersion difficult meteorological conditions could result in high daily PM_{10} concentrations observed in the southern downstream stations (Querol et al., 2009).

Figure 2b shows the daily variations in PM_{10} at different air monitoring stations in Taiwan (<http://taqm.epa.gov.tw/dust/zh-tw/Database.aspx>). It shows a clear decrease

in PM₁₀ concentrations from North Taiwan to South Taiwan during dust storm period. A comparison of daily mean PM₁₀ concentrations from three stations in south Taiwan (Kaohsiung, Pingtung, and Dongsha) shows a dramatic discrepancy in daily PM₁₀ concentration between Kaohsiung and Pingtung on 22nd March. The arrival time of the dust storm indicates that the Pingtung station was influenced by the air parcel from eastern Taiwan, which resulted in a low PM₁₀ concentration in Pingtung. Pingtung, and Dongsha showed similar low daily mean PM₁₀ concentrations, but Dongsha showed a higher maximum hourly PM₁₀ concentration (557 $\mu\text{g m}^{-3}$) when compared to Pingtung (132 $\mu\text{g m}^{-3}$). This might be because Dongsha received the high dust air parcel from South China and Pingtung received the low dust air parcel from the east coast.

In the case of metallic elements, a comparison of concentrations between this study and others (at Kaohsiung, Hengchun, Xiamen, and Gwangju) that examined the same severe ADS event in 2010 is listed in Table 3. The concentration of most elements in TSP were lower in Kaohsiung than in Xiamen, except for V, Ni, Na, Al, and Cr. Higher concentrations of some crustal elements (Mn, Co, Sr, Ba, Fe, and Mg) and some anthropogenic elements (As, Zn, and Pb) occurred in Xiamen than in Kaohsiung. The concentration ratios of As and Pb (anthropogenic elements) in Xiamen/Kaohsiung were 2.0 and 4.6, respectively (Table3), with As and Pb in Xiamen perhaps coming from coal burning (Lee et al., 2009; Zhao et al., 2011; Li et al., 2013). The higher concentrations of V and Ni in Kaohsiung may be due to heavy oil used in nearby Kaohsiung harbor shipping activities (Mueller et al., 2011; Zhao et al., 2013). These two elements have been used as markers for shipping emissions (Viana et al., 2008).

The comparison of metallic concentrations in PM₁₀ between Gwangju (Korea) and

Kaohsiung indicated that the amounts of crustal elements (Al, Ca, and Fe) collected were lower in Gwangju than Kaohsiung, but higher concentrations of Zn and Pb were observed in Gwangju. These results may reflect the difference in transport pathway and transport distance to the sites between Kaohsiung and Gwangju, in addition to local pollutant contributions. Compared with Hengchun, the mean metallic concentrations were all higher in Kaohsiung than in Hengchun, except for Na in PM_{2.5}. The concentrations of Al, Ca, Fe, Mg, and Na during ADS periods were significantly higher in Kaohsiung than in Hengchun. These concentration differences among the major elements could be due to Hengchun receiving a lower PM₁₀ concentration air parcel from the dust storm than was received by Kaohsiung, as well as local environmental conditions at the sampling site.

4. Conclusion

In this study, PM_{2.5}, PM_{2.5-10}, PM₁₀, and TSP were simultaneously collected in the Kaohsiung coastal area of southern Taiwan from February to March 2010, to distinguish the chemical characteristics and particulate profiles of NDS and ADS periods. Dust storm events were identified by back trajectory analysis, temporal and spatial variations of PM₁₀ concentrations, size ratio (PM_{2.5} and PM_{2.5-10} in PM₁₀), and Al concentrations in particles. Among the four particle fractions examined, the most striking variation in chemical characteristics between ADS and NDS periods was observed in PM_{2.5-10}. High ADS/NDS ratios for Ca, Al, Fe, Mg, Sr, Co, Ba, and Mn were observed in PM_{2.5}, PM_{2.5-10}, PM₁₀, and TSP, suggesting that these elements can also be used as indicators to identify ADS events in the study area. The variation in metallic element concentrations and EF_c values revealed that substantial crustal

elements are present in PM during ADS periods. In addition, a significant difference between the EF_c for anthropogenic elements was found in $PM_{2.5-10}$, PM_{10} , and TSP, which means that levels of these elements could be used as indicators of dust storm invasion. The spatial variations of PM_{10} concentrations also indicate that transport pathways, residence time, and topographic features were important factors affecting air quality in regions downstream of ADS source areas.

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Table 1. Ambient PM concentrations ($\mu\text{g m}^{-3}$) and ratios in the southern coastal area of Taiwan during the sampling periods.

	Date	PM _{2.5}	PM _{2.5-10}	PM ₁₀	TSP	PM _{2.5-10} /PM ₁₀	PM ₁₀ /TSP
NDS	08-09 Feb	84.9	23.4	108	180	0.22	0.60
	09-10 Feb	33.2	15.3	48.5	65.1	0.32	0.75
	22-23 Feb	41.0	14.1	55.1	72.5	0.26	0.76
	23-24 Feb	42.9	13.2	56.1	74.7	0.24	0.75
	27-28 Feb	28.3	7.9	36.2	65.1	0.22	0.56
	28-01 Feb	32.7	10.9	43.6	63.6	0.25	0.69
	09-10 Mar	47.2	24.6	71.8	134	0.34	0.54
	12-13 Mar	76.1	26.2	102	136	0.26	0.75
	15-16 Mar	33.9	14.6	48.5	68.2	0.30	0.71
	26-27 Mar	40.0	36.3	76.3	155	0.48	0.49
	Mean $\pm$ SD	46.0 $\pm$ 19.1	18.7 $\pm$ 8.7	64.7 $\pm$ 24.6	101 $\pm$ 45	0.29 $\pm$ 0.08	0.66 $\pm$ 0.10
ADS	16-17 Mar	48.5	58.3	107	202	0.55	0.53
	21-22 Mar	93.2	249	342	629	0.73	0.54
	22-23 Mar	38.7	98.6	137	243	0.72	0.57
	Mean $\pm$ SD	60.1 $\pm$ 29.1	135 $\pm$ 101	195 $\pm$ 101	358 $\pm$ 101	0.66 $\pm$ 0.10	0.55 $\pm$ 0.02

Table 2. Elemental concentrations (ng m^{-3}) in $\text{PM}_{2.5}$, $\text{PM}_{2.5-10}$, PM_{10} and TSP during the NDS^a and ADS^b periods.

Elements	$\text{PM}_{2.5}$ (mean $\pm$ SD)		$\text{PM}_{2.5-10}$ (mean $\pm$ SD)		PM_{10} (mean $\pm$ SD)		TSP (mean $\pm$ SD)	
	NDS	ADS	NDS	ADS	NDS	ADS	NDS	ADS
PM	46.0 $\pm$ 19.1	60.1 $\pm$ 29.1	18.7 $\pm$ 8.7	135 $\pm$ 101	64.7 $\pm$ 24.6	195 $\pm$ 128	101 $\pm$ 44.8	358 $\pm$ 236
V	15.2 $\pm$ 12.5	13.2 $\pm$ 5.7	2.90 $\pm$ 1.70	9.92 $\pm$ 5.41	18.1 $\pm$ 14.1	23.1 $\pm$ 11.0	24.2 $\pm$ 17.3	35.3 $\pm$ 15.2
Cr	5.67 $\pm$ 2.54	9.62 $\pm$ 5.64	4.58 $\pm$ 2.01	10.5 $\pm$ 3.3	10.3 $\pm$ 4.3	20.1 $\pm$ 9.0	14.6 $\pm$ 6.7	40.0 $\pm$ 14.1
Mn	15.5 $\pm$ 8.5	61.4 $\pm$ 54.3	10.7 $\pm$ 7.0	97.9 $\pm$ 59.6	26.2 $\pm$ 15.0	159 $\pm$ 112	45.9 $\pm$ 20.8	255 $\pm$ 184
Co	0.28 $\pm$ 0.13	0.89 $\pm$ 0.56	0.21 $\pm$ 0.12	1.49 $\pm$ 0.82	0.49 $\pm$ 0.22	2.38 $\pm$ 1.36	0.88 $\pm$ 0.41	4.76 $\pm$ 2.92
Ni	10.2 $\pm$ 7.1	9.42 $\pm$ 1.92	5.11 $\pm$ 2.17	8.59 $\pm$ 0.79	15.3 $\pm$ 9.1	18.0 $\pm$ 2.5	17.0 $\pm$ 8.4	33.6 $\pm$ 11.6
Cu	16.6 $\pm$ 4.7	17.7 $\pm$ 2.8	6.36 $\pm$ 2.30	6.35 $\pm$ 0.93	23.0 $\pm$ 6.7	24.0 $\pm$ 2.4	198 $\pm$ 132	145 $\pm$ 42
Zn	69.8 $\pm$ 41.6	94.3 $\pm$ 19.5	33.0 $\pm$ 23.1	25.4 $\pm$ 7.32	103 $\pm$ 62	120 $\pm$ 22	203 $\pm$ 87	247 $\pm$ 60
As	1.31 $\pm$ 0.53	2.19 $\pm$ 1.35	0.24 $\pm$ 0.13	1.07 $\pm$ 0.70	1.56 $\pm$ 0.64	3.26 $\pm$ 2.04	2.73 $\pm$ 1.22	7.68 $\pm$ 4.76
Se	1.40 $\pm$ 0.79	1.51 $\pm$ 0.80	0.19 $\pm$ 0.08	0.32 $\pm$ 0.16	1.58 $\pm$ 0.87	1.83 $\pm$ 0.94	3.14 $\pm$ 1.24	5.35 $\pm$ 1.84
Sr	1.59 $\pm$ 0.63	6.02 $\pm$ 5.32	2.04 $\pm$ 1.09	20.6 $\pm$ 16.3	3.63 $\pm$ 1.29	26.6 $\pm$ 21.6	9.93 $\pm$ 5.42	71.7 $\pm$ 60.3
Cd	0.45 $\pm$ 0.22	0.68 $\pm$ 0.30	0.09 $\pm$ 0.04	0.10 $\pm$ 0.02	0.54 $\pm$ 0.26	0.78 $\pm$ 0.32	1.17 $\pm$ 0.48	1.64 $\pm$ 0.39
Ba	9.33 $\pm$ 4.32	21.8 $\pm$ 15.1	8.84 $\pm$ 3.84	64.7 $\pm$ 52.2	18.2 $\pm$ 6.7	86.4 $\pm$ 67.2	28.2 $\pm$ 11.2	142 $\pm$ 95
Pb	30.3 $\pm$ 12.9	57.8 $\pm$ 27.5	5.88 $\pm$ 2.56	15.9 $\pm$ 7.0	36.2 $\pm$ 15.3	73.7 $\pm$ 34.4	54.1 $\pm$ 18.7	100 $\pm$ 38
Al	635 $\pm$ 240	2750 $\pm$ 2310	612 $\pm$ 376	8110 $\pm$ 6690	1250 $\pm$ 572	10860 $\pm$ 9000	1600 $\pm$ 1360	17200 $\pm$ 13080
Ca	117 $\pm$ 78	843 $\pm$ 686	247 $\pm$ 182	4910 $\pm$ 4570	364 $\pm$ 237	5760 $\pm$ 5250	1131 $\pm$ 782	11390 $\pm$ 10790
Fe	242 $\pm$ 121	1140 $\pm$ 1010	326 $\pm$ 181	4170 $\pm$ 3140	569 $\pm$ 276	5310 $\pm$ 4110	1480 $\pm$ 878	10560 $\pm$ 7570
Mg	152 $\pm$ 54	779 $\pm$ 605	176 $\pm$ 81	2100 $\pm$ 1590	329 $\pm$ 116	2880 $\pm$ 2180	850 $\pm$ 573	6060 $\pm$ 4580
Na	523 $\pm$ 154	728 $\pm$ 427	1040 $\pm$ 493	3670 $\pm$ 1790	1560 $\pm$ 595	4390 $\pm$ 2220	4240 $\pm$ 3020	10980 $\pm$ 5310

a: non-dust storm

b: Asian dust storm

Table.3. Comparison of metallic concentrations in aerosol collected in Kaohsiung, Hengchun, Xiamen and Gwangju during the severe ADS period (March 19-23), 2010 (unit: ng m⁻³).

Elements	Kaohsiung ^a				Hengchun ^b		Xiamen ^c	Gwangju ^d
	TSP	PM ₁₀	PM _{2.5-10}	PM _{2.5}	PM _{2.5-10}	PM _{2.5}	TSP	PM ₁₀
V	43.3	29.0	12.6	16.4	NA	NA	40.7	NA
Cr	44.6	22.9	11.6	11.3	1.83	1.81	23.0	40.0
Mn	331	205	126	78.4	5.05	5.03	612	NA
Co	5.95	2.96	1.87	1.09	NA	NA	9.19	NA
Ni	38.0	19.3	9.02	10.3	4.01	4.15	16.6	NA
Cu	167	24.7	5.82	18.8	4.39	3.86	NA	NA
Zn	233	119	21.7	97.3	7.42	6.44	327	1490
As	9.46	3.98	1.37	2.62	NA	NA	19.2	NA
Se	5.80	2.03	0.38	1.65	NA	NA	NA	NA
Sr	94.0	36.0	27.9	8.09	2.56	1.75	204	NA
Cd	1.79	0.86	0.01	0.76	0.07	0.07	NA	NA
Ba	180	113	85.2	27.5	2.17	1.78	306	NA
Pb	104	77.8	17.4	60.4	2.45	2.42	475	290
Al	22520	14640	10970	3666	433	308	21840	3630
Ca	15579	8155	6978	1177	693	376	NA	4030
Fe	13690	7231	5715	1515	312	209	19280	6300
Mg	7139	3886	2876	1009	268	211	35250	NA
Na	7704	4940	4103	837	1609	1011	3220	NA

NA: not available

a: This study (sampling period: 15:00 March 21 to 15:00 March 23, 2010)

b: Tsai et al., 2012 (sampling period: 09:00 March 21 to 09:00 March 23, 2010)

c: Zhao et al., 2011 (sampling period: 08:00-16:00 (day) and 20:00-04:00 (night) March 21-23, 2010)

d: Kwak et al., 2012 (sampling period: 14:00 March 20 to 00:00 March 21, 2010)

FIGURES

Figure 1. Location of the sampling site at NSYSU, Kaohsiung.

(a)

(b)

Figure 2. Daily variations of PM₁₀ concentrations at different air monitoring stations in (a) Eastern Asia and (b) Taiwan during the period of a severe dust storm (March 2010).

Figure 3. ADS/NDS elemental concentration ratios in PM_{2.5}, PM_{2.5-10}, PM₁₀ and TSP.

Figure 4. Average enrichment factors of metallic elements in (a) PM_{2.5}, (b) PM_{2.5-10}, (c) PM₁₀ and (d) TSP during NDS and ADS periods. (Blank on the top of bar : no significant difference; *: t-test, P<0.05; **: t-test, P<0.01).