Influences of natural and anthropogenic particles on ambient particulate air quality during typhoon season: From Bashi Channel to Kaoping River Valley

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1 | INTRODUCTION

Aeolian dust episode (ADE) is an emerging disaster occurred from the bare lands of the Kaoping River Valley in southern Taiwan because of typhoons. Four manual sampling sites located along the Kaoping River Valley conducted to collect PM$_{10}$ (aerodynamic diameter ≤ 10 μm) with high-volume samplers during the ADE occurred by Typhoon Dokson, and on regular days. Mass percentages of sea-salt particles (SSs) in PM$_{10}$ accounted for 5.47–8.91% on regular days and 11.66–14.05% in phase II. Average mass percentage of Ca$^{2+}$ in phase I increased twice than those on regular days. Cl$^{-}$ deficit percentages were much lower during the ADE (7.37–14.13%) than on regular days (31.69–42.78%), indicating acidic particles mainly produced by chemical reactions of acidic aerosols with aeolian dust and SSs. Even alkaline aeolian dust is a dominant source of the ADE, the atmospheric particles are attributable to acidic particles in the air. Hence, anthropogenic sources play a key role for the worst air quality during typhoon season.

KEYWORDS
aeolian dust episode, particulate air quality, sea-salt particles, typhoon, water-soluble ionic species
(Zhuang et al., 1999; Varun Raj et al., 2009). Thus, SSs play an important role in islands’ air ambient quality in the atmospheric and oceanic chemistry (Tsai et al., 2012; Park et al., 2015; Chen et al., 2016).

However, few studies focused on the influence of both natural particles (i.e., AD and SSs) and anthropogenic particles during the ADE of the typhoon season in island regions. Accordingly, the present study focused on the water-soluble ionic species (WSIs) characteristics in PM$_{10}$ and ascertained how natural and anthropogenic particles influence atmospheric PM$_{10}$ from Bashi Channel to Kaoping River Valley during the ADE of Typhoon Dokuri.

2  | MATERIALS AND METHODS

Four PM$_{10}$ manual sampling sites (MS1: Kaoping River Weir Management Center; MS2: Fo-Guang-Shan Buddha Memorial Museum; MS3: Yutian Elementary School; MS4: Yu-Suei Branch Campus of Huei-Nung Elementary School) and ambient air quality monitoring stations (Linyuan [LY], Daliao [MN], Pingtung [PT], and Meinong [MN] stations) along the Kaoping River Valley are shown in Figure 1a. Ambient PM$_{10}$ was collected simultaneously in a sampling network on regular days and during the ADE. Regular sampling was conducted to collect 24-hr PM$_{10}$, starting from 0800 LST (local standard time), with high-volume samplers (TE-6070D) at four sampling sites on July 7–14, 2012. The sampling during the ADE was simultaneously conducted in two phases to collect PM$_{10}$ with high-volume samplers (TE-6070D) on June 29–30, 2012, while the trajectory of the Typhoon Dokuri passed through the Bashi Channel as shown in Figure 1b. Phase I was conducted from 0100 to 0400 LST on June 29, 2012, whereas phase II was carried out from 0400 to 0800 LST of the sequential day on June 29–30, 2012. The sampling flow rate of high-volume samplers was operated at 1.4 m$^3$/min based on the standard method of NIEA A102.12A.

WSIs were analysed for ambient PM$_{10}$ sampled on regular days and during the ADE. All collected PM$_{10}$ samples were divided into half pieces. We only need one piece of them for further analysis of WSIs. The remaining one piece was on the purpose for redoing partial experiments if we were in need. One quarter of the quartz fibre filter was added inside a 15-mL bottle made of polyethylene. Each bottle was filled with distilled de-ionized water, and then vibrated ultrasonically for 60 min. The mixed solution obtained from each bottle was filtered to avoid the column damped for extending the life span of the analytical instrument. The concentrations of major anions (i.e., fluoride [F$^-$], chloride [Cl$^-$], sulfate [SO$_4^{2-}$], and nitrate [NO$_3^{-}$]) and cations (i.e., ammonium [NH$_4^+$], potassium [K$^+$], sodium [Na$^+$], calcium [Ca$^{2+}$], and magnesium [Mg$^{2+}$]) were measured with an ion chromatography (Dionex, DX-120). The quality assurance and quality control for the analysis of the WSIs were conducted in this study. At least 10% of the samples were analysed by spiking with a known amount of WSIs to determine the recovery rates. The recovery rates varied between 96 and 103%. In addition, duplicate analysis results showed that the relative percentage differences ranged from 3 to 4% for all chemical species. The sampling and analytical procedures were similar to those described in previous studies (Tsai et al., 2010; Li et al., 2015).

3  | RESULTS AND DISCUSSION

The Taiwan Central Weather Bureau recorded the ADE occurring on June 29, 2012, while Typhoon Dokuri passed along the Bashi Channel with an anticyclone outflow circulation as shown in Figure 1b. A separate outflow caused by the typhoon entered the Kaoping River Valley through its estuary in the south because the Kaoping River flows southwards. The prevailing winds were in the range of 160–200° at the DL station and 180–300° at the MN station. The wind speeds varied in the range of 4.1–7.1 m/s at the DL station and 1.2–5.4 m/s at the MN station from 0800 to 1900 LST. Even the wind speed at the MN station was less than that at the DL station, the wind speeds measured at both stations were over the threshold wind speed (3.05 m/s) for re-suspending dust in the air (Wang et al., 2015). According to Figure 1a–c, we concluded that the variation of PM$_{10}$ at the DL station was affected by the bare lands located at the estuary of the Kaoping River while the PM$_{10}$ concentrations at the MN station was influenced by the bare lands formed along the Kaoping River. This study further summarized the monthly variation of PM$_{10}$ from 2007 to 2013, which were recorded by the four Taiwan EPA’s air quality monitoring stations located along the Kaoping River Valley as shown in Figure 2a. It indicated that the monthly averaged PM$_{10}$ concentration at the DL station was 39.8 ± 6.8 μg/m$^3$ in the typhoon season (June–October). The 24-hr PM$_{10}$ concentration showed significant temporal variation as the average of 414.5 ± 432.4 μg/m$^3$, as shown in Figure 2b, exceeding the 24-hr PM$_{10}$ ambient air quality standard (125 μg/m$^3$) by 3.3-fold at the DL station on June 29, 2012. Compared to the monthly PM$_{10}$ concentration (39.8 ± 6.8 μg/m$^3$) at the DL station on regular days, the average PM$_{10}$ concentration was over 10-fold on June 29, 2012. The results evidenced that AD was the major source to deteriorate the ambient air quality along the Kaoping River during the ADE.

Site MS1 had the highest hourly average PM$_{10}$ concentrations of 677.4 μg/m$^3$ in phase I as shown in Table 1, indicating that PM$_{10}$ concentrations rose as high as 17.8- to 30.6-fold higher than the average PM$_{10}$ concentrations on regular days (22.1–37.9 μg/m$^3$). Lower hourly average PM$_{10}$ concentrations were 216.1 μg/m$^3$ at site MS2. Hourly average PM$_{10}$ concentrations at sites MS3 and MS4 located
at the left bank of the Kaoping River Valley were in the range of 73.4 and 97.6 μg/m³. The mass percentages of WSIs to PM₁₀ in phase I were higher than those on regular days by 1.17- to 1.38-fold as shown in Figure 3, indicating that PM₁₀ in phase I was rich in moisture content from the Bashi Channel to the inland area of the Kaoping River Valley, causing in high mass percentages of WSIs in PM₁₀. Among WSI species, Na⁺ and Cl⁻ commonly recognized as tracers of SSs (Chow et al., 1996). Ca²⁺ and K⁺ are related to AD (Chen et al., 2004; Lin et al., 2005; Taiwan EPA, 2008; Tsai et al., 2012), and SO₄²⁻, NO₃⁻, and NH₄⁺ are emitted from anthropogenic sources (Chen et al., 2016). Figure 3 indicated that the mass percentages of SSs (i.e., Na⁺ and Cl⁻) and AD species (i.e., Ca²⁺ and K⁺) varied greatly between the periods of regular days and phase I. The average mass percentages of 2.87 and 3.29% for Na⁺ and Cl⁻ rose more than twice to 4.34 and 6.93% in phase I. Additionally, AD species of Ca²⁺ and Mg²⁺ also increased significantly from mass percentages of 4.34–6.93% and 2.18–3.11% in phase I. However, as the Typhoon Doksur passed through the Bashi Channel in phase II, the mass percentages of Ca²⁺ dropped to the range of 2.70–3.71% and far lower than those in phase I, but still relatively higher than that on regular days. Similar to Ca²⁺, the SSs of Na⁺ and Cl⁻ also decreased to 2.06–3.48% and 2.70–5.11%, respectively. These proved that the SSs and AD increased in phase I due to the influence of Typhoon Doksur to the Kaoping River Valley. However, as Typhoon Doksur moved away from Taiwan Island, the surface wind speeds in the Valley decreased gradually, the amounts of SSs and AD were then reduced correspondingly. Moreover, a significant diurnal variation was also found for SO₄²⁻ and NO₃⁻ during the ADE. In phase I, the average mass percentages were 13.81 and 9.43% for SO₄²⁻ and NO₃⁻ and then decreased to 12.13 and 7.67% in phase II, which was possibly related to the formation of secondary inorganic aerosols in the daytime resulting from the heterogeneous reactions of SO₂ and NOₓ with AD during the transporting process (Varun Raj et al., 2009). The main chemical pathways involve the gas-phase photochemical reactions with solar radiation and oxidants such as O₃ and OH/C₁, causing SO₂ and NOₓ to form sulphate and nitrate correspondingly. High RHs and excess ammonium facilitate the transformation of gaseous SO₂ and NOₓ in aqueous phase to form sulphate and nitrate (Huang et al., 2014). In this study, meteorological data were monitored at the DL and PT air quality monitoring stations close to the sampling sites. During the ADE periods, the RHs were 75.6 ± 5.99% and
74.1 ± 9.6%, respectively, with no rainfall, indicating that the RHs were relatively high, which were favourable for the formation of WSIs from gaseous precursors. Moreover, ambient RHs were higher than 62% of deliquescence RH for ammonium sulphate and 60% of ammonium chloride (Hu et al., 2011), indicating that sulphate and chloride did exist in the aqueous phase, which favour the absorption of gaseous precursors to the aqueous phase (Sun et al., 2015).

The blowing AD was supposed to be a mixture of natural and anthropogenic particles mainly emitted from various sources along the Kaoping River Valley. Tsai et al. (2010) reported that SSs are one of the abundant atmospheric aerosols at coastal regions in southern Taiwan. We estimated the amounts of SSs using Equation (1), which is appropriate for investigating the proportions of SSs in PM$_{10}$ (Quinn et al., 2003):

$$\text{Sea salts particles} = 1.47 \times \frac{\text{Na}^+ + \text{K}^+ + \text{Mg}^{2+} + \text{Ca}^{2+} + \text{SO}_4^{2-} + \text{HCO}_3^-}{\text{Na}^+}, \quad (1)$$

where 1.47 is the mass ratio of (Na$^+$ + K$^+$ + Mg$^{2+}$ + Ca$^{2+}$ + SO$_4^{2-}$ + HCO$_3^-$)/Na$^+$. Equation (1) revealed that, in
phase I, the mass percentages of SSs to PM$_{10}$ concentrations accounting for 11.66–14.05% were obviously higher than those on regular days (5.47–8.91%) and in phase II (5.74–9.89%). The results are summarized in Table 1 that evaluated using Equation (1) was reasonable for explaining that the relatively higher SSs in PM$_{10}$ during the ADE. As AD accompanied by typhoon outflow circulation, it could bring huge amounts of SSs from the surface of the Bashi Channel to Kaoping River Valley (Park et al., 2004; Tsai et al., 2010).

SSs played as nuclei for the adsorption of sulphur dioxide and the deposition of sulphate, which could strongly enhance the oxidative capability of aerosol particles because of the release of halogen radicals in the marine surroundings (Park et al., 2015). However, Cl$^-$ is often partially depleted because of its reactions with acidic compounds such as sulphuric and nitric acids (Tsai et al., 2010). Previous studies (Quinn et al., 2003; Xu et al., 2013; Spada et al., 2015) indicated that the estimated SSs are based on the assumption that Na$^+$ and Cl$^-$ are mainly derived from seawater and hence excluding the contribution from non-sea-salt ionic species (nss-WSIs) such as K$^+$, Mg$^{2+}$, Ca$^{2+}$, SO$_4^{2-}$, and HCO$_3^-$, thereby allowing Cl loss from SSs through chemical reactions with acidic constituents of ambient particulate matter. The studied area is not far from ocean and the prevailing winds blown from Bashi Channel could enter the Kaoping River Valley from its estuary during the ADE. To reflect the actual ambient conditions, the favourable chemical reactions among different species of the WSIs should be further considered for enhancing the accuracy of estimation processes. The concentrations of nss-K$^+$, nss-Mg$^{2+}$, nss-Ca$^{2+}$, and nss-SO$_4^{2-}$ could be estimated using Equations (2)–(5) (Park et al., 2004; Kumar et al., 2012),

\[
\text{nss-K}^+ = K^+ - 0.038 \times \text{Na}^+, \tag{2}
\]
\[
\text{nss-Mg}^{2+} = Mg^{2+} - 0.12 \times \text{Na}^+, \tag{3}
\]
\[
\text{nss-Ca}^{2+} = Ca^{2+} - 0.038 \times \text{Na}^+, \tag{4}
\]
\[
\text{nss-SO}_4^{2-} = SO_4^{2-} - 0.251 \times \text{Na}^+. \tag{5}
\]

Zhuang et al. (1999) reported that the amount of nitrate associated with soil particles ([NO$_3^-$]$_{soil}$) could be estimated by subtracting the amount of nitrate associated with SSs ([NO$_3^-$]$_{ss}$) from the total nitrate ([NO$_3^-$]$_{total}$). Assuming that chloride deficit is caused entirely by both nitrate and excess sulphate, the amount of nitrate associated with SSs equals the amount of depleted chloride minus the amount of sulphate formed on SSs. The amount of nss-[NO$_3^-$] can thus be determined using Equation (6) (Zhuang et al., 1999; Yao et al., 2003),

\[
\text{nss-NO}_3^- = [\text{NO}_3^-]_{\text{total}} - [\text{NO}_3^-]_{\text{ss}} = [\text{NO}_3^-] - (1.174 \times [\text{Na}^+] - [\text{Cl}^-]), \tag{6}
\]

the mass ratio of [Cl$^-$] over [Na$^+$] (i.e., [Cl$^-$]/[Na$^+$]) in the seawater was 1.174, while their ratios in aerosol particles are always less than 1.174. Thus, the original [Cl$^-$] concentration contributed from the seawater can be estimated by 1.174* [Na$^+$]. Figure 4 demonstrates that the mass fractions of ss-WSIs and nss-WSIs in PM$_{10}$ and their spatiotemporal variations estimated on regular days and during the ADE. The major nss-WSIs and ss-WSIs in PM$_{10}$ were anthropogenic...
sources (i.e., nss-SO\(_4^{2-}\) and nss-NO\(_3^-\)) and natural sources (i.e., nss-Ca\(^{2+}\), Cl\(^-\), and Na\(^+\)). The contributions of nss-SO\(_4^{2-}\) and nss-NO\(_3^-\) in PM\(_{10}\) were much higher than those were of ss-WSIs to PM\(_{10}\) in the Kaoping River Valley. In phase I, the mass percentages of nss-WSIs in PM\(_{10}\) ranked in the order of nss-SO\(_4^{2-}\) (12.04–13.09%) > nss-NO\(_3^-\) (7.26–8.91%) > nss-Ca\(^{2+}\) (4.20–6.78%) > NH\(_4^+\) (2.57–3.07%) > nss-Mg\(^{2+}\) (1.78–3.26%) > nss-K\(^+\) (1.40–2.46%), whereas the mass percentages of ss-WSIs for PM\(_{10}\) ranked in the order of ss-Cl\(^-\) (5.00–6.87%) > Na\(^+\) (3.25–4.48%) > ss-NO\(_3^-\) (1.13–1.84%) > ss-SO\(_4^{2-}\) (0.97–1.12%) > ss-Mg\(^{2+}\) (0.46–0.54%) > ss-K\(^+\) (0.15–0.17%) > ss-Ca\(^{2+}\) (0.15–0.17%). Anthropogenic sources were substantial contributors not only on regular days but also during the ADE. The mass ratios of...
nss-SO$_4^{2-}$ to total SO$_4^{2-}$ (i.e., sum of nss-SO$_4^{2-}$ and ss-SO$_4^{2-}$) (91.9–93.0%) in phase I were slightly lower to those on the regular days (92.1–93.7%) and in phase II (92.9–95.9%). According to the concentrations of SO$_2$ and NO$_x$ monitored at the DL and PT stations ranged from 2 to 15 ppb and from 5 to 21 ppb, respectively, during the ADE periods as shown in Figure 5, highly nss-SO$_4^{2-}$ of PM$_{10}$ could be supposed to originate from anthropogenic sources near the Kaoping River Valley. High SO$_2$ and NO$_x$ concentrations remained along the Kaoping River Valley during the ADE mainly because the topography of Kaoping River that is located near the foot of the Central Range reduces wind speed as the air passes across the mountains, and consequently causes the accumulation of gaseous precursors. Moreover, Kaoping river valley is close to several industrial complexes in Kaohsiung City, which has major heavy industries, including petroleum refinery, petrochemical industry, iron works, shipbuilding industry, coal-fired power plants, municipal solid waste incinerators, etc. Large amounts of air pollutants could be emitted and transported to the nearby areas, which could cause the ambient air quality much worse than other cities in Taiwan. Therefore, SO$_2$ and NO$_x$ emitted from those major heavy industries can transfer to the concentrations of SO$_2$ and NO$_x$ monitored at the DL station. nss-Ca$^{2+}$ and nss-Mg$^{2+}$ might easily react with nss-SO$_4^{2-}$ due to the chemical acid–base neutralization reactions. This phenomenon has been observed during the ADE, indicating the amounts of crust-related constituents (such as CaSO$_4$, Ca(NO$_3$)$_2$, and Mg(NO$_3$)$_2$) in particle matter noticeably increased during the ADE since the increased nss-Ca$^{2+}$ and nss-Mg$^{2+}$ might easily react with nss-SO$_4^{2-}$ and nss-NO$_3^{-}$. Even alkaline AD is dominant source of the ADE, the atmospheric particles are attributable to acidic particles in the atmosphere. Hence, anthropogenic sources play key roles in the worst air quality in the typhoon season. In developing control strategies for the ADE, local governments need strictly regulate industries or anthropogenic activities during the ADE and take cost-effective measures for curbing the bare lands of the estuary before the typhoon season.

Chen et al. (2016) reported that the acid–base neutralization reactions were responsible for most of the chloride deficit. The Cl$^{-}$ deficit reactions could reach 100% if enough reaction time and precursors are available. The deficit of Cl$^{-}$ could be determined using Equation (7) (Quinn et al., 2003; Chen et al., 2016),

$$\text{Cl}^{-} \text{ deficit} (\%) = \frac{[\text{Cl}^{-}]_{\text{original}} - [\text{Cl}^{-}]_{\text{meas}}}{[\text{Cl}^{-}]_{\text{original}}} \times 100\%$$

$$= \frac{1.8 \times [\text{Na}^{+}]_{\text{meas}} - [\text{Cl}^{-}]_{\text{meas}}}{1.8 \times [\text{Na}^{+}]_{\text{meas}}} \times 100\%,$$

where $1.8 \times [\text{Na}^{+}]_{\text{meas}}$ is the Cl$^{-}$ concentration estimated from SSs, any loss of Cl$^{-}$ or all Na$^{+}$ in the aerosol particles is of SSs origin, [Cl$^{-}]_{\text{meas}}$ is the Cl$^{-}$ concentration measured in the aerosol particles. Table 1 illustrates the Cl$^{-}$ deficit estimated using Equation (7) in the present study. Compared with the Cl$^{-}$ deficit, their values ranged from 31.69 to 40.38% on regular days, from 7.37 to 14.13% in phase I, and from 9.97 to 27.39% in phase II. Kumar et al. (2012) reported that the deficit of Cl$^{-}$ with respect to Na$^{+}$ was mainly caused by the reaction of Cl$^{-}$ in marine particles with other acidic species (i.e., NO$_3^{-}$ and SO$_4^{2-}$) to form NaNO$_3$ and Na$_2$SO$_4$, respectively, and thus reduced the Cl$^{-}$ concentration in the atmospheric particles (Zhuang et al., 1999; Park et al., 2015). This is one of the reasons why the concentrations of nss-NO$_3^{-}$ and nss-SO$_4^{2-}$ in PM$_{10}$ during the ADE were higher than those of PM$_{10}$ on regular days. Much lower Cl$^{-}$ deficit was observed during the ADE since huge amounts of AD and SSs persistently reacting with acidic aerosols, emitted from the anthropogenic activities. Thus, the Cl$^{-}$ deficit phenomenon could be regarded as valuable references for differentiating the influence of AD and SSs in PM$_{10}$ on regular days and during the ADE. Though Cl$^{-}$ in atmospheric particles originated from a variety of anthropogenic sources such as open burning of agricultural waste (Lin et al., 2012), Pingtung County Government Environmental Bureau (2013) reported that biomass burning mostly occurs in winter and early spring due to the needs for shifting cultivation. The occurring periods of ADE and biomass burning are obviously different along Kaoping River Valley. Biomass burning should not be an important source of Cl$^{-}$ in PM during the ADE in the typhoon season. Consequently, air flow accompanied by a substantial amount of SSs resulted in a low Cl$^{-}$ deficit for the AD under the effects of strong surface winds exerted by typhoon outflow circulations.

4 | SUMMARY

A unique ADE occurred in Kaoping River Valley during the period of Typhoon Dokkiri was firstly observed in Taiwan. Anthropogenic sources close to Kaoping River still play an important role in the ambient air quality during the ADE as those in regular periods, even though the sea salts and AD were dominant natural sources accompanied by outflow circulation of Typhoon Dokkiri entering into the Kaoping River Valley during the ADE. Furthermore, chloride deficit is worthy of being investigated due to the inter-reactions derived by SSs and AD in the Kaoping River Valley in the short-term transporting process during the ADE.
ACKNOWLEDGEMENTS
This study was performed under the auspices of National Sun Yat-sen University. The authors would like to express their sincere appreciation to the Kaoping River Valley Weir Management Center, the Fo-Guang-Shan Buddha Memorial Museum, the Yutian Elementary School, and the Sui-Branch of Huei-Nung Elementary School for providing constant assistance in the field sampling of PM$_{10}$ in this study.

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How to cite this article: Lu C-C, Shen H-Z, Li T-C, Yuan C-S. Influences of natural and anthropogenic particles on ambient particulate air quality during typhoon season: From Bashi Channel to Kaoping River Valley. Atmos Sci Lett. 2018;19:e819. https://doi.org/10.1002/asl.819