How Aeolian Dust Deteriorated Ambient Particulate Air Quality along an Expansive River Valley in Southern Taiwan -A Case Study of Typhoon Dorsuki?

Supplementary Information

Chun-Chung Lu[1], Tsung-Chang Li[1], Chung-Shin Yuan[1*]

1Institute of Environmental Engineering, National Sun Yat-Sen University.

No. 70, Lian-Hai Road, Kaohsiung 804, Taiwan, R.O.C.

[*]Corresponding author:Tel: 886-7-5252000 Ext. 4409; Fax: 886-7-52524409;E-mail: ycsngi@mail.nsysu.edu.tw (C. S. Yuan)

BACKGROUND INFORMATION OF AMBIENTAIR QUALITY STATUS AND SAMPLING SITES IN THE KAOPING RIVER VALLEY

Ambient Particulate Air Quality Status

This present study summarizes the monthly PM_{10} variations from 2007 to 2013 as Fig. S1, which are recorded by the TEPA's Air Quality Monitoring Stations (AQMSs) located along the Kaoping River. The monthly average PM_{10} concentration (39.8 μg m^{-3} ± 6.8 μg m^{-3}) in typhoon season (June–October) was much lower than those on the average of non-typhoon season (78.3μg m^{-3} ± 24.1 μg m^{-3}) at the Daliao AQMS. The result evidences that the local PM_{10} contributors are not major sources to deteriorate the air ambient air quality in the Kaoping River Valley. Therefore, ADEs occurred from the topsoil at the estuary of Kaoping River should be one potential contributor for the worst air ambient quality, especially for typhoon periods.

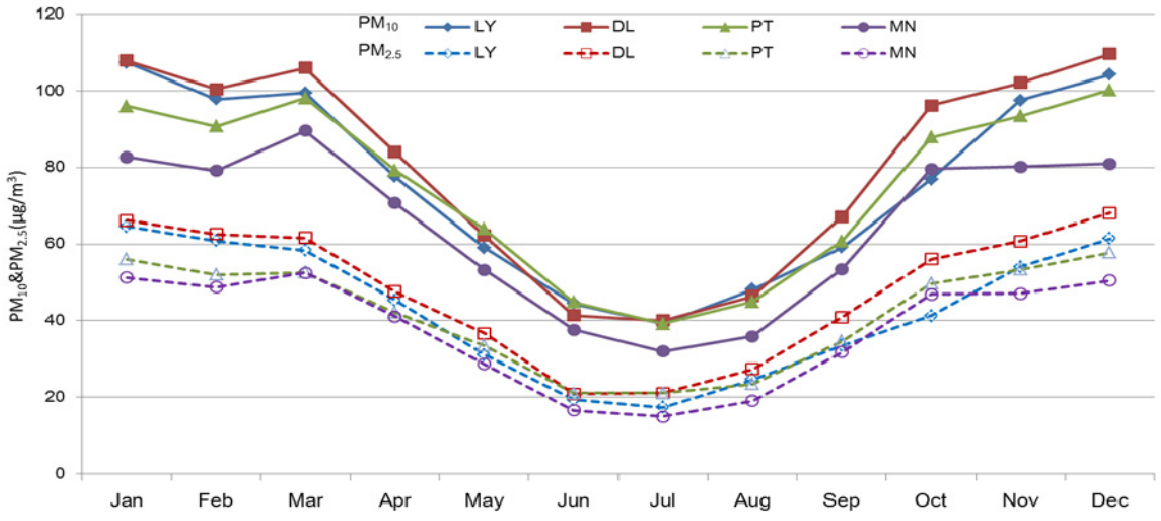


Figure S1. Monthly average PM_{10} and $PM_{2.5}$ concentrations recorded at four (LY:Linyuan, DL:Daliao, PT:Pingtung, and MN:Meinong) air quality monitoring stations in the years of 2007-2013.

Description of Sampling Sites at Right Bank

Two sampling sites were located at the right bank of the Kaoping River Valley, including the Kaoping River Weir Management Center (Site KA) (120°25'N, 22°40'E) and the Fo-Guang-Shan Buddha Memorial Museum (Site KB) (120°26'N, 22°44'E). Site KA neighboring to large exposed lands of the Kaoping River (within 5 meters) and Road No. 21 are likely influenced by the emissions from the bare lands of the riverbed and vehicular exhausts. The Fo-Guang-Shan Buddha Memorial Museum (Site KB) located at a hill is one of the most attractive scenic spots in Kaohsiung City. Site KB neighboring a parking lot can be highly influenced by the emissions from vehicular exhausts. The linear distance between Site KB and the Kapoing River course is approximately 1 km.

Description of Sampling Sites at Left Bank

Other two sampling sites, Yutian Elementary School (Site PC) (120°29'N, 22°46'E) and the Yu-Suei Branch Campus of Huei-Nung Elementary School (Site PD) (120°28'N, 22°42'E), are located at the upstream of the left bank of the Kaoping River. Site PC with an approximate distance of 1.5-2 km to the river course is located at the Ligang Township where two branches of the Kaopoing River converge together, which thus deposits fine sands to form huge bare lands with a relatively lower water level during the drought season. Site PD is located at farmlands, with a distance of

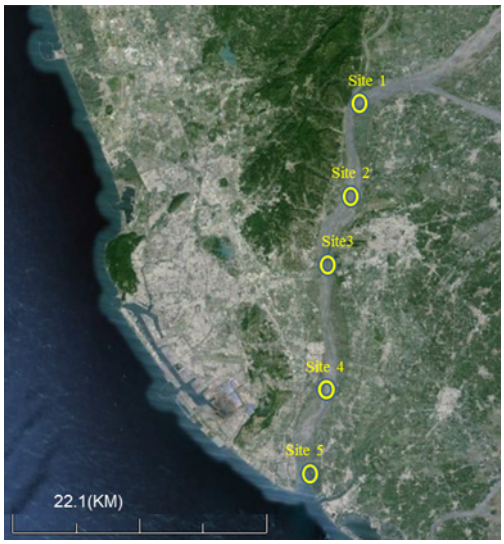
approximately 3.5 km to the river course, where agricultural residues were usually burned openly during the harvest season.

EXPERIMENTAL METHODS

Chemical fingerprints of topsoil along the riverbed

To determine the amount of ambient suspended particles contributed from the bare areas along the Kaoping River, the Chemical Mass Balance (CMB) receptor modeling is widely applied for many previous studies (Gebhart et al., 2011; Gertler et al., 2012; Sattler et al., 2012; Vega et al., 2011). We selected five representative sites at the bare lands over the riverbeds to collect topsoil samples shown in Fig. S2 (a). The soil samples were initially sieved with Tyler 400 mesh (d_p<38μm) to meet the requirement for at least 5 g and re-suspended with a dry powder atomizer in a self-designed resuspension chamber (1 m*1 m*2 m) as shown in Fig. S2 (b). Topsoil samples with aerodynamic diameters less than 10 μm (PM_{10}) were divided into fine and coarse fractions (i.e. $PM_{2.5}$ and $PM_{2.5-10}$), using a virtual impactor with a 10 μm separator at the inlet of a dichotomous sampler (Li et al., 2013a, 2013b; Tsai et al., 2011; Yuan et al., 2006). The chemical composition of PM_{10} resuspended from topsoil was then used as the source profile of aeolian dust to resolve the source apportionment of PM_{10} by using chemical mass balance (CMB) receptor model.

(a)



(b)

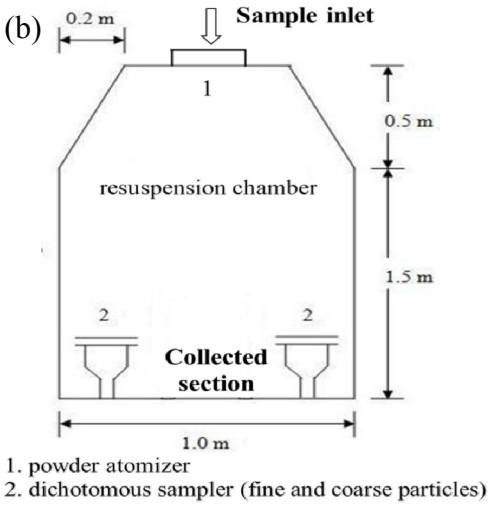


Figure S2 (a) Location of sites for collecting topsoil samples along Kaoping River. (b) Resuspension chamber used for collecting re-suspended fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) particles.

Quality assurance and quality control

Quality assurance and quality control (QA/QC) for both PM_{10} sampling and chemical analysis were conducted in this study. High-volume samplers of same brand name (TISCH Model TE-6070-HVS) were used to collect PM_{10} with the sampling flow rate of 1.4 m^3/min. Prior to conducting PM_{10} sampling, the flow rate of each PM_{10} sampler was calibrated with an orifice calibrator (X-calibrator high-volume air sampler calibrator TE-HVC-101). This sampling method was complied with the Standard Method of NIEA A102.12A. Quartz fiber filters manufactured by Pall Corporation were used in the present study. Quartz fiber filters were selected in this study since we are interested in the chemical composition of water-soluble ionic species, metallic elements, and carbonaceous content. Since silicon (Si) was not analyzed in the study. Silicon (Si) was estimated by multiplying Al by a factor of 3.41 (Hueglin et al. 2005), because it was considered in the analytical limitation of quartz fiber filter in this study. Before weighing, the quartz fiber filters were equilibrated in a

desiccator at temperatures between 20℃ and 25℃ and relative humidity (RH) between 35% and 45% for forty–eight hours. After conditioning, the filters were then weighed by an ananlytical microbalance with the precision of 1 μg to determine the PM_{10} mass. The moisture could be mostly removed in the process of conditioning (Cheng and Tsai, 2000; Yuan *et al.*, 2006). The quartz fiber filters were handled with care, so as to prevent potential cracking during the sampling procedures, as they were placed on the PM_{10} samplers. After sampling, aluminum foil was used to fold the quartz fiber filters, which were then temporarily stored at 4°C and transported back to the Air Pollution Laboratory for further chemical analysis. Both field and transportation blanks were undertaken for PM_{10} sampling, while reagent and filter blanks were applied for chemical analysis. 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 field samples. The background interference was supposed to be ignored in the present study. The sampling and analytical procedures were similar to those described in previous studies (Cheng and Tsai, 2000; Yuan et al., 2006; Tsai et al., 2008; Tsai et al., 2011; Li et al., 2013a; Li et al., 2015).

Chemical Mass Balanced (CMB) Receptor Model

Previous studies indicated that a receptor model (i.e. CMB8) could resolve the source apportionment of atmospheric particles. It activates the source profiles of prominent sources to evaluate specific contributions to a specific receptor (Kothai et al., 2008; Tsai et al., 2008). The CMB receptor model evaluates the concentrations of

different chemical components sampled at the receptor areas as a linear sum of products of source profile abundances and source contributions. For the mass conservation of each chemical species, we hypothesize a linear summation with individual contributions from various sources. Based on the function of the CMB receptor model, an "effective variance least squares" solution elevates the measurement precision for both source profiles and ambient PM_{10} concentration, providing quantitative uncertainty estimates for source apportionment. While source profiles are too similar to be distinguished from each other, the resolutions could cause large uncertainties for source contribution. These solutions also decrease the inter disturbance of chemical or physical properties in the ambient concentrations and source profiles which are possible with uncertainties or source variability. Besides, the CMB receptor model does not require background information such as meteorological conditions or emission inventories. Even though a large number of elements and chemical components are applied to differentiate among source categories, only a few apparently distinct source types are necessary to identify most of the PM_{10} in the ambient atmosphere (Chow and Watson, 2002). The concentration of species at a receptor site, C_i, can be expressed as eq 1,

$$C_i = \sum_{j=1}^{p} \alpha_{ij} F_{ij} S_j \qquad i = 1,2,3..n \tag{1}$$

where C_i is the concentration of element i of atmospheric particulate matter measured at the receptor site (μg/m^3), α_{ij} is the coefficient of element i from the source j, F_{ij} is the concentration of element i of emitted particulate matter measured at the source j (μg/m^3), and S_j is the contribution ratio of particulate matter emitted from the source j. The number i of chemical species must be no less than the number p of sources for a unique solution.

Some hypotheses which are regulated in the CMB receptor model include: (1) all chemical specie are independent and do not react with each other; (2) the constituent of source emissions is constant over the period of ambient and source sampling; (3) the number of species is greater than or equal to the number of sources; (4) all sources with a potential for contributing to the receptor have been identified and their emissions characterized; (5) the measurement uncertainties are random, uncorrelated, and normally distributed; and (6) the source profiles are linearly independent of each other.

In the present study, the ambient PM_{10} samples are averaged into representative source profiles based on data obtained from the researchers studied the chemical component of PM_{10} from local prominent sources in Taiwan and those reported by USEPA, Southern California Air Quality Study. Table 4 shows the source profile which has broadly applied for apportioning the sources of particulate matter in previous studies by the Air Pollution Laboratory in the Institute of Environmental Engineering at National Sun Yat-Sen University (Li et al., 2013a, 2013b, 2015). To

understand the source identification of the dust samples, the chemical compositions and established markers were included for further analysis.

Table S1 Source profiles of PM_{10} used for CMB receptor model

	Code	Source Profiles	References
SCT004	PBPRI1	Petroleum cracking Plant	U.S. EPA. 1991
SCT007	PP004	Industrial Boilers (Oil)	Cheng *et al.*, 2000
SCT008	PP005	Industrial Boilers (Coal)	Cheng *et al.*, 2000
SCT009	PETRO1	Petroleum Industry	U.S. EPA. 1991
SCT010	STEEL1	Steel Industry	Chiang *et al.*, 1993
SCT011	STEEL2	Coke Plant	Chiang *et al.*, 1993
SCT012	STEEL3	Sinter Plant	Chiang *et al.*, 1993
SCT013	STEEL4	Electric Arc Furnace	Yuan *et al.*, 2003
SCT020	CEMENT	Cement Industry	Chiang *et al.*, 1993
SCT023	VEHICLE2	Vehicular Exhausts	J.C Chow. 1991
SCT024	VEHICLE3	Diesel Exhausts	J.C Chow. 1991
SCT025	DUST1	Paved Road dust in South Taiwan	Cheng *et al.*, 1998
SCT026	DUST2	Paved Road dust in Central Taiwan	Cheng *et al.*, 1998
SCT027	DUST3	Paved Road dust in South Taiwan	Yuan *et al.*, 1991
SCT028	DUST4	Paved Road dust in Central Taiwan	Chiang *et al.*, 1993
SCT029	DUST5	Unpaved Road dust in Central Taiwan	Chiang *et al.*, 1993
SCT031	SOIL1	Soil Dust	U.S. EPA. 1991
SCT033	MARIN1	Marine in Central Taiwan	Cheng *et al.*, 1998
SCT034	MARIN2	Marine in South Taiwan	Chen *et al.*, 1998
SCT035	VB001	Biomass Burning	Cheng *et al.*, 1999
SCT037	SO4	Secondary Sulfate	Wang *et al.*, 2006
SCT038	NO2	Secondary Nitrate	Wang *et al.*, 2006
SCT039	Aeolian Dust	Aeolian Dust of the Kaoping River	This study

Notes:

[a] The source profiles used in this study were mainly obtained from the researcher's findings of the chemical composition of PM_{10} emitted from various emission sources. Only limited source profiles are referred from USEPA and Southern California Air Quality Study, and local emission source profiles.

Thus, twenty-two chemical species are considered as variables in the fitting species. While running CMB receptor model, five parameters should be regulated as below: (1)

the T-statistics is the proportion of the source contribution estimate to the standard error (>2.0); (2) the R^2 represents the portion of the variance in the measured concentrations that is explained by the variance calculated in the concentrations of species (between 0 and 1); (3) correlation coefficient is greater than 0.6 for a passable result; (4) chi-square (χ^2) is the weighed sum of squares of differences between the estimated and the measured fitting species concentrations (>4); (5) the estimated mass percentage is the percentage of measured PM_{10} mass concentration (between 80 and 120%).

REFERENCES

Cheng, M.T., and Tsai, Y.I. (2000). Characterization of visibility and atmospheric aerosols in urban, suburban, and remote areas. *Sci. Total Environ.* 263:101–114.

Chow, J.C., and Watson, J.G. (2002). Introduction to the A&WMA 2002 critical review- visibility: science and regulation. *J. Air Waste Manag. Assoc.* 52:626–627.

Gebhart, K.A., W.C. Malm, and M. Flores. 2011. A preliminary look at source-receptor relationships in the Texas-Mexico Border Area. *J. Air&Waste Manage. Assoc*. 50:858-868.

Gertler, A.W., Lowenthal, D.A. and Coulombe, W.G. (2012). PM_{10} source apportionment study in Bullhead City, Arizona. *J. Air&Waste Manage.* Assoc.

45:75–82.

Kothai, P., Saradhi, I.V. Prathibha, P. Hopke, P.K. Pandit, G.G. and Puranik, V.D. (2008). Source apportionment of coarse and fine particulate matter at Navi Mumbai, India. *Aerosol Air Qual. Res*. 8:423–436.

Li, T.C., Yuan, C.S. Lo, K.C. Hung, C.H. Wu, S.P. and Tong, C. (2015). Seasonal variation and chemical characteristics of atmospheric particles at three islands in the Taiwan Strait. *Aerosol Air Qual. Res.* 15: 2277–2290.

Li, T.C., Chen, W.H. Yuan, C.S. Wu, S.P. and Wang, X.H. (2013a). Physicochemical characteristics and source apportionment of atmospheric particles in Kinmen-Xiamen Airshed. *Aerosol Air Qual. Res.* 13:308–323.

Li, T.C., Chen, W.H. Yuan, C.S. Wu, S.P. and Wang, X.H. (2013b). Diurnal variation and chemical characteristics of atmospheric aerosol particles and their source fingerprints at Xiamen Bay. *Aerosol Air Qual. Res.* 13:596–607.

Sattler, M.L., and H.M. Liljestrand. 2012. Chemical mass balance model with fractionation for apportioning $PM_{2.5}$: A test case for Los Angeles Traffic Sources. *J. Air&Waste Manage. Assoc.* 55:1335–1344.

Tsai, H.H., Yuan, C.S. Hung, C.H. and Lin, Y.C. (2011). Comparing physicochemical properties of ambient particulate matter of hot spots in a highly polluted air quality zone. *Aerosol Air Qual. Res.* 10:331–344.

Tsai, H.H., Ti, T.H. Yuan, C.S. Hung, C.H. and Lin, C. (2008). Effects of sea-land breezes on the spatial and temporal distribution of gaseous air pollutants around the coastal region of Southern Taiwan. *J. of Environ. Eng. Manage.* 18:387–396.

Vega, E., I. García, D. Apam, M.E. Ruíz and M. Barbiaux. 2011. Application of a chemical mass balance receptor model to respirable particulate matter in Mexico City. *J. Air&Waste Manage. Assoc.* 61:1131–1149.

Yuan, C.S., C.C. Sau, M.C. Chen, M.H. Hung, S.W. Chang and Y.C. Lin. 2004. Mass concentration and size-resolved chemical composition of atmospheric aerosols sampled at pescadores islands during Asian Dust Storm periods in the years of 2001 and 2002. TAO, 15:857–879.

Yuan, C.S., Hai, C.X. and Zhao, M. (2006). Source profiles and fingerprints of fine and coarse sands resuspended from soils sampled in Central Inner Mongolia. *China Part.* 4:304–311.