Available online at www.sciencedirect.com
ScienceDirect
Journal of Hazardous Materials 157 (2008) 418–422
Journal of Hazardous Materials
www.elsevier.com/locate/jhazmat

Source characterization of total suspended particulate matter near a riverbed in Central Taiwan

Chi-Wen Lin*, Ju-Fang Yeh, Tsang-Chih Kao

Department of Environmental Engineering, Da-Yeh University, No. 112, Shanjiao Road, Dacun, Changhua, 51591, Taiwan, ROC

Received 25 September 2007; received in revised form 30 December 2007; accepted 4 January 2008
Available online 12 January 2008

Abstract

Samples of total suspended particulate (TSP) matter were collected by using TSP samplers from certain areas representing the estuary of the Jhuoshuei River, Taiwan during monsoon and non-monsoon seasons in 2005. A total of 12 elements and nine inorganic ions were identified by inductively coupled plasma atomic emission spectrometry (ICP-AES) and ion chromatography (IC), respectively. Enrichment factors explaining a preponderance of the variance in the data were applied to the data sets. The results show that wind direction significantly affected the concentration of TSP during the monsoon season; moreover, these concentrations were also markedly higher during the monsoon than during the non-monsoon season. Low enrichment factor (EF) values (1.18–2.88) were observed during the monsoon for Ca, Fe, Na, Ba, Cd, Co, Li, Mn, and Sr, reflecting the importance of dust contribution by natural processes. Conversely, the EF values calculated for Ca, K, Ba, Co, Li, and Zn were relatively high (7.03–20.26) when observed during non-monsoon season, a phenomenon suggesting that they are mainly contributed from non-crustal sources. The observations of relatively enhanced EF values during monsoon indicate that the changes in wind direction between monsoon and non-monsoon seasons are associated with the different contribution sources. The high concentration of TSPs observed in the ambient air is believed to be primarily due to surface soil particle emission from the riverbeds.

Keywords: Chemical species; Enrichment factors (EF); Monsoon season; Total suspended particulate (TSP)

1. Introduction

Particulate matter may be generated by natural processes or through human activities, including soil dust, diesel trucks, power plants, wood stoves, and industrial processes [1–4]. Individual particles vary considerably in size, geometry, chemical composition, and physical properties. Ambient particulate matter is one of six criteria pollutants, of which the effects on human health and the environment vary with the physical and chemical composition of each.

Total suspended particulate (TSP) matter has received worldwide attention, due to its potential impact on global climate change and its effects on human health worldwide. Many governments and international organizations have set guideline levels for average ambient particulate matter concentration. In 1987, the US EPA replaced the TSP standard with a National Ambient Air Quality Standard (NAAQS) for PM_{10} in response to research showing that these smaller particles present a greater health risk due to their ability to penetrate deeper into the lungs. According to the Taiwanese EPA air quality standards in an ambient environment, the annual and daily average thresholds of PM_{10} are 65 and 125 $\mu g\,m^{-3}$, respectively; the daily average threshold of TSPs is 250 $\mu g\,m^{-3}$. TSP concentrations remain elevated, despite the great efforts exerted by the Taiwanese government towards reduction by controlling the emission rates of local pollutant sources.

The downstream extremities of the Jhuoshuei River are heavily deposited with fine soil particles due to poor geological conditions in the watersheds and concentrated rainfall during the summer season. These deposits are difficult to improve; hence they easily become a source of dust particle emissions during the dry winter monsoon. There are certain indications that the southwestern riverbank area of the Jhuoshuei River (i.e., Yunlin County) might receive elevated TSP matter via fugitive dust emission from the riverbed [5].

In this study, we investigated the significance of TSP matter as the cause of the air pollution in Yunlin County because large quantities of soil particles are known to be emitted from the

* Corresponding author. Tel.: +886 4 851 1339; fax: +886 4 851 1336.
E-mail address: cwlin@mail.dyu.edu.tw (C.-W. Lin).

0304-3894/$ – see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2008.01.015

riverbed of the Jhuoshei River. Whether the monsoon season pronouncedly affects the TSP concentration was also studied. A calculation of enrichment factors was performed for all elements in the experimental data sets to identify the emission sources of TSP and the effects of the monsoon on the TSP concentration in Yunlin County.

2. Materials and methods

2.1. Site descriptions

Changhua County, located in the west-central region of Taiwan, has a population of 1.3 million people and covers an area of approximately 1074 km^2. Surrounded by mountains to the north, south, and east, the county faces the Taiwan Strait on the west. Yunlin County, having an area of about 1290 km^2, is located in the south-central portion of western Taiwan and is at the northern tip of Chianan Plain. Constituting the longest river in Taiwan, the Jhuoshuei River originates from a stream on the highest peak of Mt. Hohuan in Nantou County and a subsidiary stream 187 km in length. The administrative regions through which the stream passes include Nantou, Changhua, and Yunlin Counties. In the near-coastal areas, the river separates Yunlin County from Changhua County. The Formosa Industrial Park, which contributes 1500 tonnes of TSP per year, is situated near the southwestern section of the river. Measurements were conducted at seven locations within the estuary in Changhua and Yunlin Counties, Taiwan, as shown in Fig. 1. Site no. 1 (Dacheng), called the upwind site during the monsoon season, is located in Changhua County, being situated on the northern bank of the Jhuoshuei River. Site numbers 2, 3, 4, and 5, designated downwind sites, are Mailiao-SC, Lunbei-FN, Mailiao-SJ, and Lunbei-FR, respectively. These sites are located in Yunlin County, being situated on the southern bank of the river. The number 6 and 7 sites are surface-soil sampling sites located on the dry riverbed.

2.2. Sampling program

The collection of TSP samples was implemented during both monsoon (11/4N–11/10N; 11/28D–12/4N; D represents daytime; N represents nighttime) and non-monsoon seasons (7/30N–8/3D; 8/26N–8/30D; 9/20D–9/26D) at five sites (sites nos. 1–5) in both Changhua and Yunlin Counties. The air flow rate was controlled at 1.2 m^3 min^{-1} and the sampling was conducted over a 12 h period, either from 08:00 to 20:00 h or 20:00 to 08:00 h. A total of 202 samples were collected by using a TSP sampler (121 FT, Kimoto, Japan), each sample being collected on a 20 cm × 25 cm quartz filter. To measure the TSP concentration, all filters were weighed before and after sampling. All procedures were strictly quality-controlled to avoid any adverse effects on the samples. Detailed procedures regarding operating conditions for samplers and QA/QC procedures can be found in our previous publication [6]. The samplers were located in positions that were 10–15 m higher than ground level where neither obstacles nor emission sources such as incinerators or stacks were situated nearby.

2.3. Chemical analysis

2.3.1. Elemental analysis

In our study, inductively coupled plasma atomic emission spectrometry (ICP-AES, Optimal 2000, MA) was used to identify a total of 12 elements (Al, Ca, Fe, K, Na, Ba, Cd, Co, Li, Mn, Sr, and Zn). The instrument operating conditions were set according to the information recommended on the case for the device. Certified standards (1000 mg l^{-1}; Merck) were used for the determination of trace element concentrations in the samples. Due to the sensitivity of ICP, all samples and blanks were digested in closed beakers for 6–8 h at 75–85 °C with the addition of a 19 ml acid mixture consisting of boric, nitric, hydrochloric, and hydrofluoric acids with proportions of 10:5:2:2, respectively. After particles in the filter were completely dissolved in the mixed acid solution, the new solution was filtered, after which 50 ml of nitric acid was added (0.5N) to decrease the acidity below 2%.

2.3.2. Ion analysis

Ion chromatography (IC) (model DX 120, Dionex, CA) is a high-performance version of ion-exchange chromatography that has become the preferred method for routine analysis. This method has typically been applied in cation and anion analy-

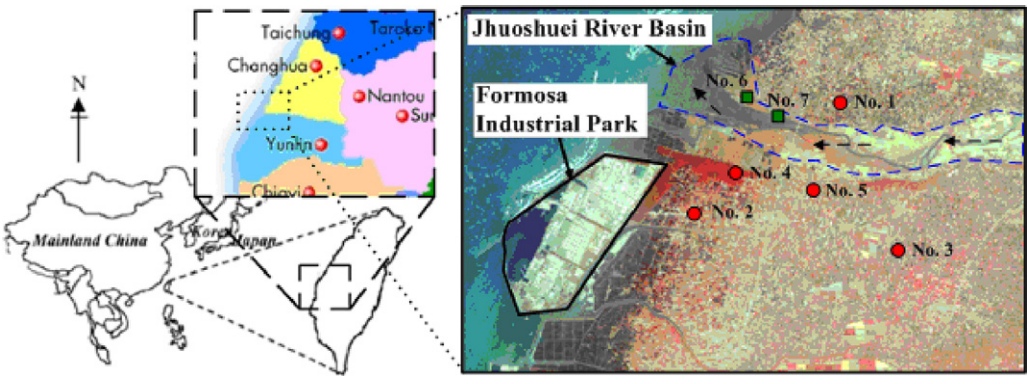

Fig. 1. Location of aerosol sampling sites in estuary of Jhuoshuei River, Taiwan. Numbers 1, 2, 3, 4, 5, 6, and 7 represent sampling sites at Dacheng, Mailiao-SC, Lunbei-FN, Mailiao-FJ, Lunbei-FR, on-site soil sampling 1, and on-site soil sampling 2, respectively.

Table 1
TSP concentration during monsoon and non-monsoon seasons

Sites	Monsoon season		Non-monsoon season	
	Sample count	Mean (standard deviation)	Sample count	Mean (standard deviation)
No. 1	27[b]	129.74 (54.14)	29[c]	90.61 (41.34)
No. 2	27[b]	191.40 (187.12)	29[c]	98.16 (36.19)
No. 3	27[b]	203.28 (156.29)	29[c]	118.80 (48.95)
No. 4	7[d]	190.84 (119.05)	7[e]	119.99 (36.48)
No. 5	7[d]	192.65 (31.27)	7[e]	82.60 (20.32)
No. 5	6[f]	813.43 (881.26)	0	–
Downwind sites[a]	68[a]	194.54[a]	72	104.89

Concentration unit: $\mu g\,m^{-3}$.
[a] "Downwind sites" include four sites (nos. 2–5), excluding data collected during 12/2D–12/4N.
[b] Sampling during monsoon seasons (11/4N–11/10N; 11/28D–12/4N).
[c] Sampling during non-monsoon seasons (7/30N–8/3D; 8/26N–8/30D; 9/20D–9/26D).
[d] Sampling during 11/7N–11/10N (monsoon).
[e] Sampling during 9/23D–9/26D (non-monsoon).
[f] Sampling during 12/2D–12/4N (monsoon).

ses in many previous studies [7,8]. In our study, nine inorganic ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_2^-, NO_3^-, and SO_4^{2-}) were analyzed by IC. Before analyzing the ion compositions, the solid suspended particulate matter in the filter paper was removed into the extracting liquid by ultrasonic shake extraction. In the composition analysis of anions, 9 mM Na_2CO_3 was used as the eluent to drive the samples at a flow rate of 1.0 ml min^{-1}.

2.4. Enrichment factors

Analysis of the enrichment factor (EF) was introduced in the 1970s. This method, an indirect tool, can be utilized to differentiate between metallic pollution originating from human activities and from natural processes and to assess the degree of anthropogenic influence [9–12]. The formula to calculate EFs can be generalized as Chester and Stoner proposed in 1973 [13]:

$$EF_{(El)_{crust}} = \frac{[El]_{sample}/[X]_{sample}}{[El]_{crust}/[X]_{crust}} \quad (1)$$

where (El) is the concentration of the chemical element under consideration, and (X) is the concentration of the reference element [14]. Aluminum is frequently used as a reference element on the basis of the assumption that its anthropogenic sources to the atmosphere are negligible [8,15–17].

If the value of EF approaches 1, the earth's crust is the predominant source. EF values of chemical elements in the range of 1–5 suggest no significant contribution of anthropogenic sources to the ambient level of these elements. Operationally, when given the local variation in soil composition, an EF value greater than 5 suggests that a significant portion of the element has been contributed from non-crustal sources [18]. As the EF value increases, the non-crustal-source contribution also increases. EF values of chemical elements higher than 10 indicate anthropogenic-source origins [1,4,19].

3. Results and discussion

3.1. Spatial variations of particulate matter components

Table 1 indicates that the average TSP concentrations are in the range of 129.7–813.43 $\mu g\,m^{-3}$ during the monsoon and 82.6–119.9 $\mu g\,m^{-3}$ during the non-monsoon season. It appears that the average TSP concentration during the monsoon is generally higher than during the non-monsoon season. The highest TSP concentration observed during December 2–4 is attributed to the strong wind during monsoon. In addition, the average TSP concentration is the lowest at no. 1 (129.74 $\mu g\,m^{-3}$) and the highest at no. 5 (813.43 $\mu g\,m^{-3}$) during the monsoon. The apparent difference in concentration between these two sites is believed to be site-specific, no. 1 being upwind and no. 5 downwind. In contrast, the average TSP concentration is lowest at no. 5 (82.6 $\mu g\,m^{-3}$) and highest at no. 4 (119.9 $\mu g\,m^{-3}$) during the non-monsoon season. Moreover, the wind direction is not stable during the non-monsoon season; however, it is quite stable and primarily from north to south during the monsoon. This observation suggests that the wind direction significantly affects the TSP concentration at each site.

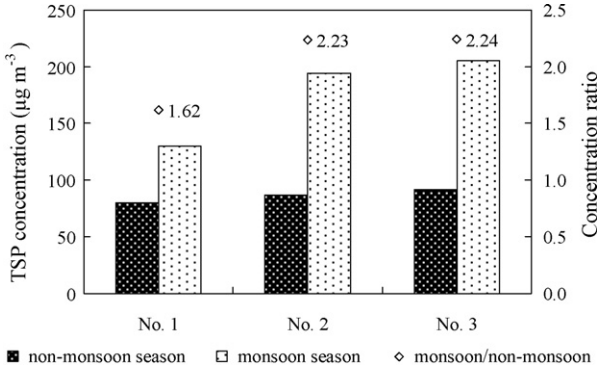

Fig. 2. TSP concentration during monsoon and non-monsoon seasons.

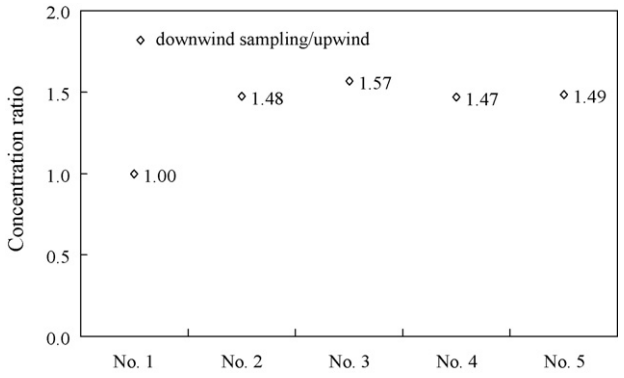

Fig. 3. Comparison of TSP concentration for downwind and upwind sampling during monsoon season.

Fig. 2 indicates that during the non-monsoon season, the TSP concentration at no. 4 and no. 3 is quite similar to that at the no. 1 site (98.1 and 118.8 $\mu g\,m^{-3}$, respectively, compared with 90.6 $\mu g\,m^{-3}$). Furthermore, the average TSP concentration at all sites during the monsoon is significantly higher than during the non-monsoon season. Specifically, the no. 1, no. 2, and no. 3 sites are 1.62, 2.23, and 2.24 times higher, respectively. Moreover, Fig. 3 shows that during the monsoon, the average TSP concentration at the downwind sites (nos. 2–5) is significantly higher than at the upwind site (no. 1), i.e., no. 2, no. 3, no. 4, and no. 5 sites are approximately 50% higher, respectively. Consequently, we suggest that the existing sources are located between the upwind and downwind sites. Moreover, these sources significantly affect TSP concentration at the downwind sites during the monsoon.

3.2. Identification of emission sources via enrichment factors

In this study, several chemical elements and Al, which is used for calculating EF values, were extracted from the surface soil in the riverbed of the Jhuoshuei River. Fig. 4 shows that the EF values for the elements (Ca, Fe, Na, Ba, Co, Cd, Li, Mn, and Sr) sampled during the monsoon at five sampling sites range from 1 to 5, except for K and Zn. These values imply that there is no significant contribution from other sources (e.g., anthropogenic); however, the enrichment of these elements is attributed to a soil origin (i.e., dust from the riverbed) during the monsoon.

Fig. 5 indicates that the EF values for Fe, Na, Cd, Mn, and Sr sampled during the non-monsoon season at the no. 1, no. 2 and no. 3 sites range from 1 to 5, implying that the enrichment of these elements is from a soil origin. Elements such as Ca, K, Ba, Co, Li, and Zn, with EF values greater than 5, suggest that the enrichment of these elements is mainly contributed from non-crustal sources. The EF values in Fig. 4 for most of the trace elements sampled during the monsoon are lower than those in Fig. 5 during the non-monsoon season. This observation indicates that the change in wind direction from monsoon to non-monsoon seasons also led to the different contribution sources.

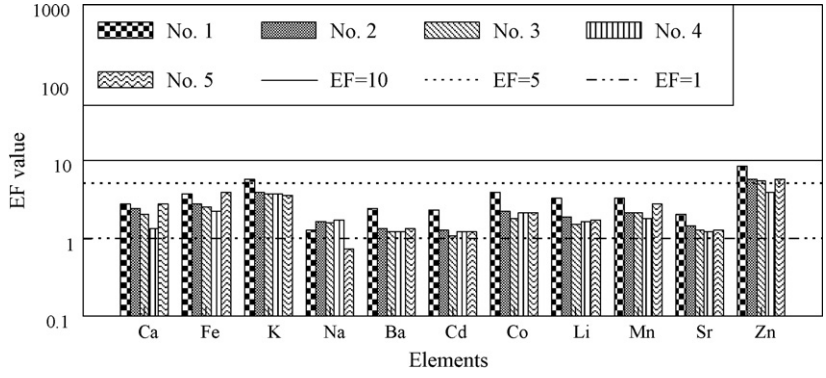

Fig. 4. Enrichment factors for elements (sampling during monsoon season).

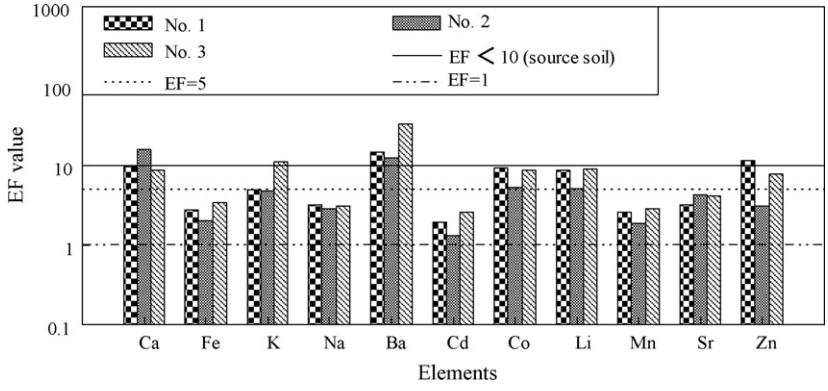

Fig. 5. Enrichment factors for elements (sampling during non-monsoon season).

On the basis of the TSP concentration analysis, we believe that certain sources markedly affect the TSP concentration in Yunlin County during the monsoon but not significantly during the non-monsoon season and that these sources are located within Changhua and Yunlin Counties. Moreover, the enrichment factors indicate that these sources are natural, being primarily derived from soil dust. The monsoon usually occurs between October and December, which is still the dry season. During this dry season, the water level in the Jhuoshuei River decreases while the dryness of the riverbed soil increases. Additionally, during the monsoon, the wind direction is usually from north to south and with significant speed. The result is that a large amount of soil dust is transported from the Jhuoshuei River to Yunlin County by the monsoon winds, thereby leading to the high concentration of TSP. It should be noted that the concentration of TSP during the non-monsoon season is insignificantly increased because the wind direction during this season is not stable.

4. Conclusion

In this study, the particulate-matter monitoring indicated that the average TSP in Yunlin County was significantly greater than the Taiwanese EPA guideline level. The average concentrations of TSP at the downwind sites during the monsoon were significantly higher than those at the upwind site; however, there was no significant difference between the average concentrations of TSP at all sampling sites during the non-monsoon season. The wind direction is frequently from north to south during the monsoon, thereby significantly affecting the TSP concentration. Fugitive dust emitted from surface soil from the Jhuoshuei River was the main cause of the elevated concentration of TSP during the monsoon.

Acknowledgements

The authors would like to thank the air-quality group (San-Ju Lu, Chih-Hsien Yu, Ming-Chieh Liu, Ting-Hsuan Chen, Yu-Hsing Yang, Chiang-Hao Lu, and Li-Hsuan Chen) at Da-Yeh University for their valuable support in field sampling and analysis. The authors also wish to express appreciation to Dr. Cheryl J. Rutledge for her editorial assistance.

References

[1] P.A. Scheff, R.A. Wadden, Toxic air pollutants in Chernivtsi, Ukraine, Environ. Int. 23 (1997) 273–290.

[2] D. Voutsa, C. Samara, T. Kouimtzis, K. Ochsenkuhn, Element composition of airborne particulate matter in the multi-impacted urban area of Thessaloniki, Greece, Atmos. Environ. 36 (2002) 4453–4462.

[3] Y. Sun, G. Zhuang, Y. Wang, L. Han, J. Guo, M. Dan, W. Zhang, Z. Wang, Z. Hao, The air-borne particulate pollution in Beijing – concentration, composition, distribution and sources, Atmos. Environ. 38 (2004) 5991–6004.

[4] J.H. Han, K.J. Moon, S.Y. Ryu, Y.J. Kim, K.D. Perry, Source estimation of anthropogenic aerosols collected by a DRUM Sampler during spring of 2002 at Gosan, Korea, Atmos. Environ. 39 (2005) 3113–3125.

[5] EPB Yunlin, State implementation plan, Environmental Protection Bureau, Yunlin County, Taiwan, ROC, 2005.

[6] C.W. Lin, R.F. Yeh, Estimating dust emission from a sandbank on the downstream Jhuoshuei River under strong wind conditions, Atmos. Environ. 41 (2007) (2007) 7553–7561.

[7] K. Ohta, K. Tanaka, Simultaneous determination of common mono- and divalent cations in natural water samples by conductimetric detection ion chromatography with an unmodified silica gel column and oxalic acid/18-crown-6 as eluent, Anal. Chim. Acta 381 (1999) 265–273.

[8] I.F. Al-momani, Trace elements in atmospheric precipitation in northern Jordan measured by ICP-MS: acidity and possible sources, Atmos. Environ. 37 (2003) 4507–4515.

[9] A.W. Struempler, Trace element composition in atmospheric particulates during 1973 and the summer of 1974 at Chadron, Nebraska, Environ. Sci. Technol. 9 (1975) 1164–1168.

[10] J.L. Moyers, L.E. Ranweller, S.B. Hopf, N.E. Korte, Evaluation of particulate trace species in southwest desert atmosphere, Environ. Sci. Technol. 11 (1977) 789–795.

[11] P. Quevauviller, Impact of industrial and mine drainage waste on the heavy metal distribution in the drainage basin and estuary of the Sado River (Portugal), Environ. Pollut. 59 (1989) 267–286.

[12] J. Isakson, M. Oblad, E.S. Lindgren, M.D. Fridell, J.M. Pacyna, M. Makinen, Perturbation of background aerosol at rural sites in the Nordic Counties, Atmos. Environ. 31 (1997) 3077–3086.

[13] R. Chester, J.H. Stoner, Average trace element composition of low level marine atmospheric particulates, Nature 246 (1973) 138–139.

[14] C. Reimann, P. Caritat, Distinguishing between natural and anthropogenic sources for elements in the environment: regional geochemical surveys versus enrichment factors, Sci. Total Environ. 337 (2005) 91–107.

[15] L. Poissant, J.P. Schmit, P. Beron, Trace inorganic elements in rainfall in the Montreal Island, Atmos. Environ. 28 (1994) 339–346.

[16] L. Gidhagen, H. Kahelin, P. Schmidt-Thome, C. Johansson, Anthropogenic and natural levels of arsenic in PM_{10} in Central and Northern Chile, Atmos. Environ. 36 (2002) 3803–3817.

[17] G. Gullu, G. Dogan, G. Tuncel, Atmospheric trace element and major ion concentrations over the Eastern Mediterranean Sea: identification of anthropogenic source regions, Atmos. Environ. 39 (2005) 6376–6387.

[18] Y. Gao, E.D. Nelson, M.P. Field, Q. Ding, H. Li, R.M. Sherrell, C.L. Gigliotti, D.A. Van Ry, T.R. Glenn, S.J. Eisenreich, Characterization of atmospheric trace elements on $PM_{2.5}$ particulate matter over the New York–New Jersey harbor estuary, Atmos. Environ. 36 (2002) 1077–1086.

[19] V.K. Mishra, K.H. Kim, C.H. Kang, K.C. Choi, Wintertime sources and distribution of airborne lead in Korea, Atmos. Environ. 38 (2004) 2653–2664.