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Measurement of Ambient Air Arsenic (As) Pollutant Concentration and Dry Deposition Fluxes in Central Taiwan

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ABSTRACT

This study investigated the feasibility of monitoring arsenic (As) levels in total suspended particulates (TSP), in both dry deposition and dry deposition flux at five sampling sites located in central Taiwan during the years of 2009–2010. Experimental results indicate that the average As concentration in TSP and dry deposition was highest in Quan-xing (industrial) and lowest in Gao-mei (wetland). The mean As composition in TSP was highest in Hei-mei (residential) and lowest in Gao-mei (wetland). The average highest arsenic (As) of seasonal concentrations and compositions in TSP, dry deposition were occurred in winter and fall denotes that fossil fuel combustion by Taichung thermal power plant (TTPP) emissions and heating by household were the main reasons responsible for the high data values measured. Atmospheric concentrations of arsenic (As) were analyzed by a ICP-MS (Perkin Elmer Sciex ELAN DRC II). Finally, best-fit model can be used successfully in the prediction of ambient air pollutants around suburban/coastal, downtown, residential, industrial and wetland areas.

Keywords: TSP; dry deposition; best-fit overall dry deposition velocity model; arsenic; ICP-MS.

INTRODUCTION

Arsenic in particulate matter poses a great threat to human health. In the second edition of the Air Quality Guidelines for Europe, published by the World Health Organization (WHO) in 2000, no amount is safe when inhaled

Arsenic gets into the atmosphere from natural and anthropogenic sources. The greatest natural emission source of atmospheric arsenic is from active volcanoes, whereas a small amount comes from vegetation and wind-driven dust. The greatest anthropogenic arsenic emission results from pyrometallurgical operations in the production of non-ferrous metals, combustion of fossil fuels and the use of pesticides. High concentrations and/or long exposure of metals may cause adverse effects on human health, even though As concentrations are a small fraction of PM (Berggren *et al*., 1990; Devries *et al*., 1996). Determination of metals composition of inhalable particles is important in determining their potential impact on human health (Allen *et al*., 2001; Park *et al*., 2008). It has been estimated that anthropogenic emission is three times higher than the natural (WHO, 2000). Coal-fuelled power plants along with copper and Pb smelters are the largest point sources of As in the air, while pesticides, fungicides, weed killers and wood treatment products also release a significant amount of As into the environment (Huang *et al*., 1994).

Previous studies on As species in total suspended particles (Oliveira *et al*., 2005) and PM_{10} (Sanchez-Rodas *et al*., 2007) in the area of Huelva have shown that the presence of As (III) is directly related to anthropogenic activities. The current investigation measures toxic (As, Cd, Cr, Hg, Ni and Pb), anthropogenic (Ba, Cu, Mn, Sb, Se, Sr, Ti, V and Zn) and crust (Al, Ca, Fe, K and Mg) elements from a 2,000 MW heavy oil-fired power plant (Wang *et al*., 2010). The results indicate that Cu, Pb, S, Zn, As, and Cd came mainly from anthropogenic sources. Results from CMB receptor model indicated that major sources in Harbin were traffic dusts, road dusts, coal burning dusts and dusts from petrochemical plants (Huang *et al*., 2010).

Arsenic exists in several states of oxidation: As (0) or as ion forms like As (V) arsenate, As (III) arsenite and As (III) arsine. It is generally recognized that the soluble inorganic arsenicals are more toxic than the organic ones, and the inorganic As (III) species are more toxic than the inorganic As (V) (Duker *et al*., 2005). The predominance of a particular wind direction determines the distribution of PM and As (Serbula *et al*., 2010). Accordingly, high dry deposition velocities are threatening to human health by dry deposition (Chang *et al*., 2003). The average

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calculated/measured mass dry deposition flux ratios value by Baklanov and Sorensen model was 4.65 while the average calculated/measured mass dry deposition flux ratios values by Zhang was 7.16. Both of these two models overestimate the measured dry deposition mass flux. The Baklanov model performed better results in the prediction of mass dry deposition flux (Fang *et al*., 2010). High concentrations were recorded in the winter seasons for both particulate pollutants. In winter season, temperature is low and wind speed is generally low (Vijay Bhaskar and Mehta, 2010). Overall, the results indicate that motor vehicles and industrial plumes are the major sources of elevated UFPs in urban/industrial air (Young and Keeler, 2007). In Auckland, as expected, road transport was considered the dominant source of PM_{10} (Senaratne *et al*., 2005). High wind speed also could have resulted in high PM_{10} and $PM_{2.5}$ levels due to the re-suspension of particulate matter under well dispersed conditions (Cheng and Li, 2010).

Recent advances have been made by using a surrogate surface to directly measure dry deposition. This technique can be used to directly assess deposited material and allows comparisons to be made between measured and modeled data (Holsen and Noll, 1992; Davidson *et al*., 1985). Measurements made with a smooth surrogate surface with a sharp leading edge have been shown to agree well with results of modeled dry deposition if complete atmospheric size distributions are used (Holsen and Noll, 1992; Paode *et al*., 1998; Sofuoglu *et al*., 1998).

This study focuses on the following objectives: 1) to measure the ambient air particulates dry deposition, the total suspended particulate (TSP) concentration and the concentration of arsenic (As) that is attracted to them; 2) to determine the ambient air particulates arsenic (As) dry deposition, concentration and deposition velocity; 3) to compare the dry deposition fluxes that are calculated using various models with measured dry deposition fluxes for arsenic (As) at five (suburban/coastal, downtown, residential, industrial and wetland) sampling sites.

METHODS

Sampling Program

Fig. 1 presents the five characteristic sampling sites. All the samples were sampling for 1380–1400 min during the sampling period for each sampling group. They are as follows.

Bei-shi sampling station (24°13'31.82"N, 120°34'09.45"E) located in Shalu, Taichung, Taiwan. This sampling station is a suburban/coastal station and its perimeter is entirely unobstructed. It is in a residential area, with a heavy traffic expressway approximately 2 km east of the station. A Taiwan EPA air-quality monitoring station is located roughly 10 m northwest of the Bei-shi sampling station.

Chang-Hua sampling station (24°05'24.52"N, 120°31' 31.73"E) is within the city, where heavy automobile emissions and emissions from a chemical plant in central Taiwan are present.

He-mei town sampling station (24°06'00.54"N, 120°30' 51.34"E) is in a typical residential area with most pollution sources associated with anthropogenic activities and vehicular emissions.

Quan-xing sampling station (24°08'37.89"N, 120°29' 09.43"E) is located in Shen-kang, a town with a total area of 246.8 hectares, of which 126.5 hectares is occupied by factories, in which industrial activities such as primary metal, textile, basic metal, machinery, petroleum and coal products are manufactured.

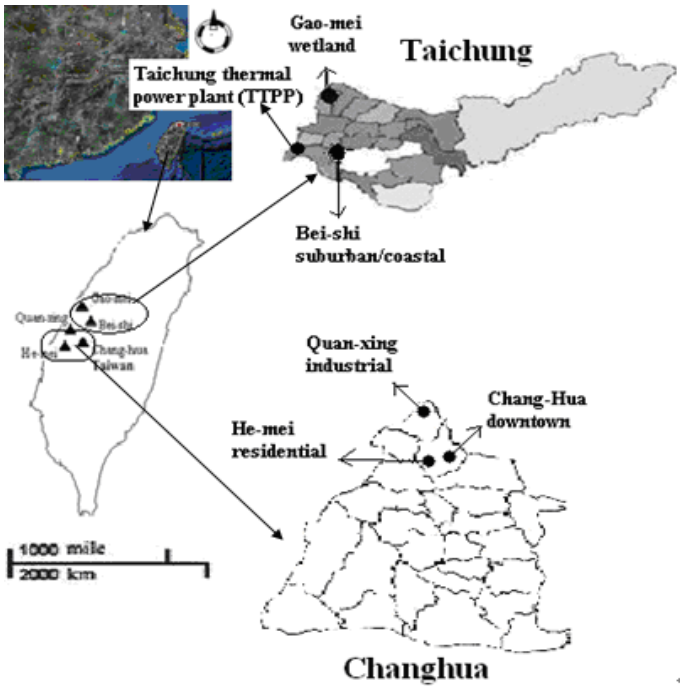

Fig. 1. Geographical location at five characteristic sampling sites in central Taiwan.

Gao-Mei wetland sampling station (24°18'35.07"N, 120°33'08.21"E) is located in Shimizu, Taichung, Taiwan. The town includes more than 300 hectares in central Taiwan. Taichung thermal power plant (TTPP) was built on 281 hectares and located on the coast on the west side of the sampling site. It is a coal combustion-based TTPP, and it supplies approximately 4,400 MW of electricity to central Taiwan.

Sampling Program

PS-1 Sampler

PS-1 collects suspended particulate matter. The maximum size of the collected particles was approximately 100 μm (Graseby-Andersen, GMW High Volume Air Sampler). The PS-1 sampler is a complete air sampling system that is designed to collect suspended airborne particles. The flow rate was adjusted to 200 L/min in this investigation. A quartz filter (diameter 10.2 cm) was used to filter the suspended particles. The filters were first conditioned for 24 hours in an electric chamber at a humidity of 35 ± 5% and a temperature of 25 ± 5°C prior to both on-and off-weighing. The filters were placed in a sealed CD box during transport and storage. The sampling device and procedures were similar to those used in our earlier study (Fang *et al*., 2009).

Dry Deposition Plate

A dry deposition plate (DDP) that consists of a smooth, horizontal, surrogate surface was adopted to estimate a lower bound on the dry deposition flux. The DDP consisted of a smooth surface plate that was made of polyvinyl chloride (PVC) and measured 21.5 cm long, 8.0 cm wide and 0.8 cm thick. The DDP also had a sharp, leading edge that pointed into the prevailing wind. All filters were maintained at 50% relative humidity and 25°C for over 48 h. Before they were used in sampling, all of the filters were weighed to an accuracy of 0.0001 g (Chu *et al.*, 2008).

Chemical Analysis

The samples were placed in an oven during the night before they were weighed. A quarter of each filter was cut and selected for use in the digestion process. 3 mL of hydrochloric acid (HCl) and 9 ml of nitrate (HNO_3) (Yong *et al*., 1992) were mixed and added to a thin digested filter during heating. The whole digestion process lasted for two hours. The filtration process proceeded after digestion. Furthermore, 0.2 % HNO_3 was added to the sampler for quantitative analysis. Measurements of the arsenic (As) were made using an ICP-MS (Perkin Elmer Sciex ELAN DRC II).

Dry Deposition Velocity Model

Best-fit Overall

Holsen *et al.* (2001) adopted the best-fit method to obtain overall dry deposition velocities of ambient air pollutants from the slope of the regression line between the measured dry deposition flux (*y*-axis) and the ambient concentration (*x*-axis). These best-fit overall dry deposition velocities were also used to obtain the calculated dry deposition flux of arsenic (As) at the five sampling sites.

Quality Control

Blank test background contamination was monitored by using operational blanks (unexposed projection film and quartz filter) which were processed simultaneously with field samples. The field blanks were exposed in the field when the field sampling box was opened to remove and replace field samples. Background contamination of heavy metals was accounted for by subtracting field blank values from the concentrations. Field blank values were very low, usually below or around the method detection limits. In this study, the background contamination is insignificant and can be ignored. The results of the blank test are 0.20μg for arsenic (As).

Recovery Efficiency Test

At least 10% of the samples are analyzed by spiking with a known amount of arsenic (As) to calculate recovery efficiencies. The analysis procedure for the recovery test is the same as that described for the field samples. The recovery percentages for arsenic (As) were 98%.

Reproducibility Test

We repeated the analysis of the same standard solution many times. The reproducibility test indicates the stability of the instruments. The relative standard deviation varied between 95% and 100%, and the mean relative deviation was smaller than 5%.

RESULTS AND DISCUSSION

Spring season covered the months from February to April. Summer season covered the months from May to July. Fall season covered the months from August to October. Winter season covered the months from November to January.

Table 1 displays the sampling information for atmospheric arsenic (As) at Bei-shi (suburban/coastal) sampling site during year 2009–2010. The results demonstrate that the average wind speeds, temperatures, relative humidity and dry deposition velocity were 1.68 (m/s), 22.73 (°C), 75.69 (%) and 1.01 (cm/s), respectively. The average monthly dry-deposited arsenic (As) order was Nov. (2.23 ng/m^2-min) > Dec. (2.22 ng/m^2-min) > Apr. (2.14 ng/m^2-min) > Oct. (2.14 ng/m^2-min) > Sep. (1.95 ng/m^2-min) > Jan. (1.88 ng/m^2-min) > May (1.42 ng/m^2-min) > Mar. (1.54 ng/m^2-min) > Feb. (1.53 ng/m^2-min) and the seasonal variations of As dry deposition were in the order fall > winter > summer > spring. The average monthly As concentrations in the total suspended particulates (TSP) order was Oct. (3.61 ng/m^3) > Jan. (3.6 ng/m^3) > Sep. (3.47 ng/m^3) > Feb. (3.42 ng/m^3) > Nov. (3.37 ng/m^3) > Apr. (3.21 ng/m^3) > Dec. (3.18 ng/m^3) > Mar. (2.97 ng/m^3) > May (2.9 ng/m^3) and the seasonal variations of As concentrations in TSP were in the order fall > winter > spring > summer. Finally, the average monthly As compositions in the total suspended particulates (TSP) order was Jan. (138.7 ng/g) > Oct. (129.6 ng/g) > Dec. (100.6 ng/g) > May (98.3 ng/g) > Feb.

Table 1. Sampling information for atmosphere pollutant arsenic (As) at Bei-shi (suburban/coastal) sampling site during year 2009–2010.

Sample group	Sample date	WS (m/sec)	Temp (°C)	RH (%)	As Dry deposition (ng/m^2-min)	As in TSP (cm/sec)	Dry deposition Velocity (cm/sec)	As in Tsp (ng/g)
Range	Sep.'09							
(n = 20)	Min	1.66	29.72	67.44	1.32	2.89	0.73	22.7
	Max	2.32	29.32	73.44	2.36	4.21	1.36	74.6
	Average	1.89	28.91	70.32	1.95	3.47	0.95	60.9
Range	Oct.'09							
(n = 20)	Min	1.21	22.36	60.8	1.78	2.98	0.83	63.9
	Max	1.78	25.28	79.24	2.7	4.22	1.14	155.8
	Average	1.48	24.56	65.99	2.12	3.61	0.98	129.6
Range	Nov.'09							
(n = 20)	Min	0.96	20.21	62.24	1.98	2.64	0.89	45.7
	Max	2.23	24.88	82.16	2.64	4.31	1.67	110.4
	Average	1.44	22.91	76.11	2.23	3.37	1.44	80.7
Range	Dec.'09							
(n = 20)	Min	1.25	17.48	64.76	1.7	2.31	0.81	77.3
	Max	1.82	20.72	81.36	2.61	4.01	1.67	133.5
	Average	1.61	19.39	70.27	2.22	3.18	1.22	100.6
Range	Jan.'09							
(n = 20)	Min	0.92	14	56.56	1.45	2.77	1.55	94.5
	Max	1.39	22	84.6	2.31	4.4	1.22	154.3
	Average	1.08	19.53	71.75	1.88	3.6	0.9	138.7
Range	Feb.'09							
(n = 20)	Min	1.06	17.64	83.36	0.99	2.64	0.55	42.5
	Max	1.86	19.76	92.2	1.87	4.58	1.13	100.8
	Average	1.55	18.67	87.5	1.53	3.42	0.77	95.3
Range	Mar.'09							
(n = 20)	Min	1.32	20.92	79.92	1.22	2.63	0.76	77.3
	Max	2.32	24.08	88.52	2.01	3.57	1.22	98.2
	Average	1.72	22.78	84.1	1.54	2.97	0.87	90.5
Range	Apr.'09							
(n = 20)	Min	1.42	16.16	76.6	1.68	2.56	0.82	40.3
	Max	2.97	27.52	87.6	2.85	4.01	1.68	89.2
	Average	2.03	22.38	79.55	2.14	3.21	1.15	75.7
Range	May'09							
(n = 20)	Min	1.22	26.96	70.96	1.42	2.42	0.8	64.3
	Max	3.16	28.52	81.28	2.21	3.21	1.4	108.2
	Average	2.29	27.63	77.51	1.83	2.9	1.06	98.3
Min		0.92	14	56.56	0.99	2.31	0.55	22.7
Max		3.16	29.72	92.2	2.85	4.58	1.68	155.8
Mean		1.68	22.73	75.69	1.94	3.3	1.01	96.7

(95.3 ng/g) > Mar. (90.5 ng/g) > Nov. (80.7 ng/g) > Apr. (75.7 ng/g) > Sep. (60.9 ng/g) and the seasonal variations of As compositions in TSP were in the order winter > summer > fall > spring at Bei-shi (suburban/coastal) sampling site.

The results also demonstrate that the average highest and lowest monthly for dry-deposited arsenic (As) were in Nov. and Feb., respectively. In addition, the average highest and lowest monthly arsenic (As) concentrations in TSP were in Oct. and May, respectively. Finally, the average highest and lowest monthly arsenic (As) compositions in TSP were in Jan. and Sep., respectively at Bei-shi (suburban/coastal) sampling site.

Table 2 displays the sampling information for atmospheric arsenic (As) at Chang-hua (downtown) sampling site during year 2009–2010. The results demonstrate that the average wind speeds, temperatures, relative humidity and dry deposition velocity were 1.65 (m/s), 23.49 (°C), 70.74 (%) and 0.96 (cm/s), respectively. The average monthly dry-deposited arsenic (As) order was Dec. (2.15 ng/m^2-min) > Apr. (2.07 ng/m^2-min) > Oct. (2 ng/m^2-min) > Sep. (1.98 ng/m^2-min) > Mar. (1.92 ng/m^2-min) > Feb., Nov. (1.79 ng/m^2-min) > Jan. (1.76 ng/m^2-min) > May (1.67 ng/m^2-min) and the seasonal variations of As dry deposition were in the

Table 2. Sampling information for atmosphere pollutant arsenic (As) at Chang-hua (downtown) sampling site during year 2009–2010.

Sample group	Sample date	WS (m/sec)	Temp (°C)	RH (%)	As Dry deposition (ng/m^2-min)	As in TSP (cm/sec)	Dry deposition Velocity (cm/sec)	As in Tsp (ng/g)
Range	Sep.'09							
(n = 20)	Min	1.04	29.44	61.2	1.67	2.64	0.84	22.7
	Max	2.72	31.56	69.84	2.15	3.87	1.06	87.9
	Average	1.57	30.5	66.83	1.98	3.32	1	56.1
Range	Oct.'09							
(n = 20)	Min	0.63	22.56	62.24	1.54	2.68	0.69	77.2
	Max	1.7	26.64	74.16	2.68	4.03	1.67	134.2
	Average	1.02	26.22	69.62	2	3.23	1.08	119.4
Range	Nov.'09							
(n = 20)	Min	0.97	23.72	60.68	1.01	2.79	0.45	55.9
	Max	1.46	26.24	76.8	2.21	3.74	1.14	95.3
	Average	1.26	25.02	70.66	1.79	3.34	0.91	74.3
Range	Dec.'09							
(n = 20)	Min	1.04	17.16	60.68	1.85	3.41	0.74	68.2
	Max	2.08	21.08	75.96	2.6	4.53	1.07	122.9
	Average	1.4	19.26	67.11	2.15	3.83	0.94	92.7
Range	Jan.'09							
(n = 20)	Min	0.88	17.04	55.72	1.18	2.98	0.54	100.8
	Max	1.81	22.64	77.44	2.03	4.02	1.11	155.2
	Average	1.23	19.12	68.21	1.76	3.68	0.81	127.7
Range	Feb.'09							
(n = 20)	Min	1.02	17.4	74.96	1.39	3.3	0.61	44.8
	Max	1.59	18.72	82.52	2.03	4.43	0.92	100.6
	Average	1.39	17.85	78.13	1.76	3.91	0.77	127.7
Range	Mar.'09							
(n = 20)	Min	1.26	21.88	70.88	1.65	2.53	0.87	53.8
	Max	2.04	25.68	77.84	2.13	3.83	1.4	99.6
	Average	1.63	24.2	74.62	1.92	3.09	1.06	83.4
Range	Apr.'09							
(n = 20)	Min	1.01	15.32	67	1.85	2.85	0.98	33.9
	Max	2.72	27.24	76.32	2.23	4.01	1.19	87.2
	Average	1.81	20.27	71.31	2.07	3.31	1.05	69.7
Range	May'09							
(n = 20)	Min	1.86	27.52	64.08	1.43	2.44	0.83	44.9
	Max	3.22	29.52	71.8	1.88	3.11	1.28	107.2
	Average	2.59	28.66	68.32	1.67	2.81	1	90.5
Min		0.63	15.32	55.72	1.01	2.44	0.45	22.7
Max		3.22	31.56	85.52	2.68	4.53	1.67	155.2
Mean		1.65	23.49	70.74	1.9	3.32	0.96	89.1

order fall > spring > winter > summer. The average monthly As concentrations in the total suspended particulates (TSP) order was Feb. (3.91 ng/m^3) > Dec. (3.83 ng/m^3) > Jan. (3.68 ng/m^3) > Nov. (3.34 ng/m^3) > Sep. (3.32 ng/m^3) > Apr. (3.31 ng/m^3) > Oct. (3.23 ng/m^3) > Mar. (3.09 ng/m^3) > May (2.81 ng/m^3) and the seasonal variations of As concentrations in TSP were in the order winter > fall > spring > summer. Finally, the average monthly As compositions in the total suspended particulates (TSP) order was Jan. (127.7 ng/g) > Oct. (119.4 ng/g) > Dec. (92.7 ng/g) > May (90.5 ng/g) > Feb. (87.8 ng/g) > Mar. (83.4 ng/g) > Nov. (74.3 ng/g) > Apr. (69.7 ng/g) > Sep. (56.1 ng/g) and the seasonal variations of As compositions in TSP were in the order winter > summer > fall > spring at Chang-hua (downtown) sampling site.

The results also demonstrate that the average highest and lowest monthly for dry-deposited arsenic (As) were in Dec. and May, respectively. In addition, the average highest and lowest monthly arsenic (As) concentrations in TSP were in Feb. and May, respectively. Finally, the average highest and lowest monthly arsenic (As) compositions in TSP were in Jan. and Sep., respectively at Chang-hua (downtown) sampling site.

Table 3 displays the sampling information for atmospheric

arsenic (As) at He-mei (residential) sampling site during year 2009–2010. The results demonstrate that the average wind speeds, temperatures, relative humidity and dry deposition velocity were 2.09 (m/s), 23.39 (°C), 73.61 (%) and 1.23 (cm/s), respectively. The average monthly dry-deposited arsenic (As) order was Oct. (2.37 ng/m^2-min) > Nov. (2.29 ng/m^2-min) > Sep. (2.23 ng/m^2-min) > Jan. (2.22 ng/m^2-min) > Dec. (2.13 ng/m^2-min) > Apr. (2.05 ng/m^2-min) > Feb. (1.92 ng/m^2-min) > May (1.91 ng/m^2-min) > Mar. (1.90 ng/m^2-min) and the seasonal variations of As dry deposition were in the order fall > winter > spring > summer. The average monthly As concentrations in the total suspended particulates (TSP) order was Dec. (3.15 ng/m^3) > Apr. (3.04 ng/m^3) > Jan. (3.03 ng/m^3) > Mar., Sep. (2.96 ng/m^3) > Feb. (2.95 ng/m^3) > Oct. (2.86 ng/m^3) > May (2.74 ng/m^3) > Nov. (2.6 ng/m^3) and the seasonal variations of As concentrations in TSP were in the order winter > fall > spring > summer. Finally, the average monthly As compositions in the total suspended particulates (TSP) order was Nov. (152.3 ng/g) > Feb. (140.3 ng/g) > Oct. (110.5 ng/g) > Dec. (107.9 ng/g) > Jan., May (99.5 ng/g) > Mar. (99.4 ng/g) > Sep. (96.4 ng/g) > Apr. (61.6 ng/g) and the seasonal variations of As compositions in TSP were in the order winter > fall > spring > summer at He-mei (residential) sampling site.

Table 3. Sampling information for atmosphere pollutant arsenic (As) at He-mei (residential) sampling site during year 2009–2010.

Sample group	Sample date	WS (m/sec)	Temp (°C)	RH (%)	As Dry deposition (ng/m^2-min)	As in TSP (cm/sec)	Dry deposition Velocity (cm/sec)	As in Tsp (ng/g)
Range	Sep.'09							
(n = 20)	Min	1.34	29.4	62.64	1.98	2.34	0.95	64.8
	Max	2.6	31.52	72.62	2.67	3.48	1.67	111.7
	Average	1.88	30.44	69.02	2.23	2.96	1.29	96.4
Range	Oct.'09							
(n = 20)	Min	1.46	25.52	62.48	2.03	2.47	1.11	43.9
	Max	2.11	26.44	75.84	2.72	3.25	1.67	138.7
	Average	1.83	26.16	71.36	2.37	2.86	1.4	110.5
Range	Nov.'09							
(n = 20)	Min	0.96	23.57	63.05	1.89	2.16	1.03	94.7
	Max	1.21	26.08	79.96	2.8	3.05	2.16	185.3
	Average	1.1	24.9	72.77	2.29	2.6	1.51	152.3
Range	Dec.'09							
(n = 20)	Min	1.72	17.5	61.52	1.9	2.77	0.95	64.7
	Max	3	23.32	78.72	2.45	3.64	1.32	125.3
	Average	2.33	19.55	69.08	2.13	3.15	1.14	107.9
Range	Jan.'09							
(n = 20)	Min	0.97	17.14	58.52	2.07	2.46	0.97	74.7
	Max	3.32	22.5	81.04	2.54	3.12	1.49	123.5
	Average	1.73	19.06	71.93	2.22	2.95	1.24	99.5
Range	Feb.'09							
(n = 20)	Min	1.36	17.64	77.96	1.66	2.64	1	100.8
	Max	2.72	18.94	85.58	2.1	3.74	1.29	169.3
	Average	2.24	18.02	81.3	1.92	3.03	1.09	140.3
Range	Mar.'09							
(n = 20)	Min	1.32	21.54	75.15	1.7	2.63	0.95	33.5
	Max	2.25	25.36	82	2.06	3.31	1.3	119.7
	Average	1.72	23.91	78.49	1.9	2.96	1.08	99.4
Range	Apr.'09							
(n = 20)	Min	2.06	15.76	70.26	1.77	2.45	0.95	22.7
	Max	4.42	26.9	78.58	2.44	3.85	1.27	79.8
	Average	3.21	20.33	73.79	2.05	3.04	1.14	61.6
Range	May'09							
(n = 20)	Min	1.7	26.96	69.72	1.65	2.42	0.96	77.9
	Max	3.43	29	77.8	2.1	2.93	1.35	118.4
	Average	2.75	28.14	73.79	1.91	2.74	1.17	99.5
Min		0.96	15.76	58.52	1.65	2.16	0.95	22.7
Max		4.42	31.52	85.58	2.8	3.85	2.16	185.3
Mean		2.09	23.39	73.61	2.11	2.92	1.23	111.0

The results also demonstrate that the average highest and lowest monthly for dry-deposited arsenic (As) were in Oct. and Mar., respectively. In addition, the average highest and lowest monthly arsenic (As) concentrations in TSP were in Dec. and Nov., respectively. Finally, the average highest and lowest monthly arsenic (As) compositions in TSP were in Nov. and Apr., respectively at He-mei (residential) sampling site.

Table 4 displays the sampling information for atmospheric arsenic (As) at Quan-xing (industrial) sampling site during year 2009–2010. The results demonstrate that the average wind speeds, temperatures, relative humidity and dry deposition velocity were 2.63 (m/s), 23.32 (°C), 76.71 (%) and 1.11 (cm/s), respectively. The average monthly dry-deposited arsenic (As) order was Nov. (2.89 ng/m^2-min) > Dec. (2.76 ng/m^2-min) > Oct. (2.61 ng/m^2-min) > Sep. (2.53 ng/m^2-min) > Apr. (2.47 ng/m^2-min) > May (2.27 ng/m^2-min) > Feb. (2.07 ng/m^2-min) > Mar. (2.06 ng/m^2-min) > Jan. (1.74 ng/m^2-min) and the seasonal variations of As dry deposition were in the order fall > summer > winter > spring. The average monthly As concentrations in the total suspended particulates (TSP) order was Jan. (3.93 ng/m^3) > Oct. (3.88 ng/m^3) > Sep. (3.85 ng/m^3) > Nov. (3.74 ng/m^3) > Feb. (3.67 ng/m^3) > Dec. (3.60 ng/m^3) > Apr. (3.48

Table 4. Sampling information for atmosphere pollutant arsenic (As) at Quan-xing (industrial) sampling site during year 2009–2010.

Sample group	Sample date	WS (m/sec)	Temp (°C)	RH (%)	As Dry deposition (ng/m^2-min)	As in TSP (cm/sec)	Dry deposition Velocity (cm/sec)	As in Tsp (ng/g)
Range	Sep.'09							
(n = 20)	Min	1.16	29.32	64.08	2.31	3.25	0.89	88.3
	Max	3.7	31.48	75.4	2.84	4.35	1.44	157.8
	Average	2.19	30.37	71.2	2.53	3.85	1.12	137.1
Range	Oct.'09							
(n = 20)	Min	1.23	25.48	62.72	2.01	3.64	0.84	44.6
	Max	3.27	26.36	77.76	3.01	4.24	1.31	133.5
	Average	2.64	26.1	73.1	2.61	3.88	1.12	100.8
Range	Nov.'09							
(n = 20)	Min	0.77	23.42	65.42	2.65	3.15	1.18	67.9
	Max	1.1	25.92	83.33	3.15	4.29	1.67	144.6
	Average	0.93	24.78	76.88	2.89	3.74	1.3	105.2
Range	Dec.'09							
(n = 20)	Min	2.34	17.84	63.36	2.34	2.98	1.2	90.4
	Max	3.93	21.56	81.48	3.18	4.18	1.48	169.3
	Average	3.25	19.84	71.05	2.76	3.6	1.28	144.9
Range	Jan.'09							
(n = 20)	Min	0.86	17.24	61.32	1.12	3.44	0.51	78.3
	Max	4.84	22.36	84.64	2.58	4.62	1.07	128.3
	Average	2.23	18.99	75.65	1.74	3.93	0.75	102.7
Range	Feb.'09							
(n = 20)	Min	1.7	17.88	80.96	1.88	3.26	084	67.9
	Max	3.87	19.16	88.64	2.34	4.04	1.13	139.2
	Average	3.1	18.19	84.47	2.07	3.67	0.95	115.5
Range	Mar.'09							
(n = 20)	Min	1.39	21.21	79.42	1.76	2.6	0.78	22.9
	Max	2.45	25.04	86.17	2.31	4.22	1.26	77.9
	Average	1.82	23.62	82.37	2.06	3.46	1.02	58.7
Range	Apr.'09							
(n = 20)	Min	2.56	16.2	72.6	2.01	2.99	0.97	55.9
	Max	6.13	25.56	81.4	2.84	4.02	1.58	142.7
	Average	4.6	20.38	76.26	2.47	3.48	1.21	124.8
Range	May'09							
(n = 20)	Min	1.54	26.4	75.36	1.99	2.77	0.87	33.9
	Max	3.65	28.48	83.8	2.55	3.85	1.46	96.9
	Average	2.91	27.62	79.36	2.27	3.2	1.22	63.7
Min		0.77	16.2	61.32	1.12	2.6	0.51	22.9
Max		6.13	31.48	88.64	3.18	4.62	1.67	169.3
Mean		2.63	23.32	76.71	2.38	3.64	1.11	105.9

ng/m^3) > Mar. (3.46 ng/m^3) > May (3.2 ng/m^3) and the seasonal variations of As concentrations in TSP were in the order fall > winter > spring > summer. Finally, the average monthly As compositions in the total suspended particulates (TSP) order was Dec. (144.9 ng/g) > Sep. (137.1 ng/g) > Apr. (124.8 ng/g) > Feb. (115.5 ng/g) > Nov. (105.2 ng/g) > Jan. (102.7 ng/g) > Oct. (100.8 ng/g) > May (63.7 ng/g) > Mar. (58.7 ng/g) and the seasonal variations of As compositions in TSP were in the order winter > fall > spring > summer at Quan-xing (industrial) sampling site.

The results also demonstrate that the average highest and lowest monthly for dry-deposited arsenic (As) were in Nov. and Jan., respectively. In addition, the average highest and lowest monthly arsenic (As) concentrations in TSP were in Jan. and May, respectively. Finally, the average highest and lowest monthly arsenic (As) compositions in TSP were in Dec. and Mar., respectively at Quan-xing (industrial) sampling site.

Table 5 displays the sampling information for atmospheric arsenic (As) at Gao-mei (wetland) sampling site during year 2009–2010. The results demonstrate that the average wind speeds, temperatures, relative humidity and dry deposition velocity were 2.16 (m/s), 22.58 (°C), 76.46 (%) and 1.11 (cm/s), respectively. The average monthly dry-deposited arsenic (As) order was Oct. (2.13 ng/m^2-min) > Nov. (2.0 ng/m^2-min) > Apr. (1.92 ng/m^2-min) > Dec. (1.89 ng/m^2-min) > Feb. (1.86 ng/m^2-min) > Mar. (18.4 ng/m^2-min) > Jan. (1.83 ng/m^2-min) > May (1.44 ng/m^2-min) and the seasonal variations of As dry deposition were in the order fall > winter > spring > summer. The average monthly As concentrations in the total suspended particulates (TSP) order was Oct. (3.11 ng/m^3) > Nov.

Table 5. Sampling information for atmosphere pollutant arsenic (As) at Gao-mei (wetland) sampling site during year 2009–2010.

Sample group	Sample date	WS (m/sec)	Temp (°C)	RH (%)	As Dry deposition (ng/m^2-min)	As in TSP (cm/sec)	Dry deposition Velocity (cm/sec)	As in Tsp (ng/g)
Range	Oct.'09							
(n = 20)	Min	1.19	25.28	64.76	1.93	2.93	1.1	40.6
	Max	2.17	26.6	76.62	2.33	3.36	1.21	80.8
	Average	1.89	26.12	73.07	2.13	3.11	1.14	72.3
Range	Nov.'09							
(n = 20)	Min	0.87	23.15	67.55	1.66	2.89	0.94	35.9
	Max	1.12	25.7	81.97	2.31	3.31	1.17	98.2
	Average	0.99	24.66	76.79	2	3.03	1.1	82.9
Range	Dec.'09							
(n = 20)	Min	1.81	17.9	62.76	1.58	1.98	1	55.8
	Max	3.2	21.54	80.36	2.1	3.05	1.77	144.7
	Average	2.46	19.85	70.55	1.89	2.72	1.2	114.2
Range	Jan.'09							
(n = 20)	Min	0.95	17.08	62.64	1.54	2.33	0.99	40.7
	Max	3.86	22.38	83.13	2.11	2.84	1.33	98.2
	Average	1.93	19.03	75.08	1.83	2.68	1.14	80.9
Range	Feb.'09							
(n = 20)	Min	1.47	18.18	79.32	1.73	1.95	1.03	66.8
	Max	2.85	19.36	87.08	1.93	3.04	1.46	123.5
	Average	2.35	18.48	82.56	1.86	2.72	1.16	105.2
Range	Mar.'09							
(n = 20)	Min	1.3	21.62	78.53	1.76	2.34	1.01	33.5
	Max	2.06	25.26	83.48	1.91	3.04	1.32	88.3
	Average	1.66	23.95	80.82	1.84	2.69	1.15	74.6
Range	Apr.'09							
(n = 20)	Min	2.03	16.42	72.2	1.65	2.78	0.89	15.8
	Max	5.02	26.62	79.52	2.35	3.22	1.14	64.7
	Average	3.52	20.68	75.49	1.92	2.99	1.08	46.2
Range	May'09							
(n = 20)	Min	1.52	26.85	74.42	1.21	2.45	0.8	20.7
	Max	3.17	28.56	80.44	1.66	3.22	1.08	88.9
	Average	2.6	27.84	77.34	1.44	2.66	0.91	70.3
Min		0.87	16.42	62.64	1.21	1.98	0.8	15.8
Max		5.02	28.56	87.08	2.35	3.36	1.77	144.7
Mean		2.16	22.58	76.46	1.86	2.83	1.11	84.3

(3.03 ng/m^3) > Apr. (2.99 ng/m^3) > Dec., Feb. (2.72 ng/m^3) > Mar. (2.69 ng/m^3) > Jan. (2.68 ng/m^3) > May (2.66 ng/m^3) and the seasonal variations of As concentrations in TSP were in the order fall > winter > spring > summer. Finally, the average monthly As compositions in the total suspended particulates (TSP) order was Dec. (144.2 ng/g) > Feb. (105.2 ng/g) > Nov. (82.9 ng/g) > Jan. (80.9 ng/g) > Mar. (74.1 ng/g) > Oct. (82.3 ng/g) > May (70.3 ng/g) > Apr. (46.2 ng/g) and the seasonal variations of As compositions in TSP were in the order winter > spring > fall > summer at Gao-mei (wetland) sampling site.

The results also demonstrate that the average highest and lowest monthly for dry-deposited arsenic (As) were in Oct. and May, respectively. In addition, the average highest and lowest monthly arsenic (As) concentrations in TSP were in Oct. and May, respectively. Finally, the average highest and lowest monthly arsenic (As) compositions in TSP were in Dec. and Apr., respectively at Gao-mei (wetland) sampling site.

The main sources for Bei-shi (suburban/coastal) sampling site was science park, fossil fuel combustion and transportation. As for Quan-xing (industrial) sampling site, the nearby source were steel industry, electronic industry, plastic industry, chemical industry, basic metal manufacturing, machinery manufacturing, petroleum and coal products. The main sources for Gao-mei (wetland) sampling site were Taichung thermal power plant (TTPP) and fossil fuel combustion. As for Chang-hua (downtown) sampling site, the nearby source were transportation, chemical plant and fossil fuel combustion. Finally, the main sources for He-mei (residential) sampling site were transportation, fossil fuel combustion, heating and waste incineration.

Table 6 displayed the comparison of ambient air As concentrations (ng/m^3) at various particle sizes for different characteristic sampling sites during year of 2000–2010. The results indicated that the maximum average particulate concentration for As in PM_{10} was occurred at China (2004) (average, 42.1 ng/m^3) (Zheng *et al*., 2004). This value was about 5.4, 4.3, 7.1, 2.5, 1.4, 1.1, 1.7, 38.3 and 6.8 times higher that of Huelva (2001) (Sanchez-Rodas *et al*., 2007), Huelva (2002) (Sanchez-Rodas *et al*., 2007), Taiwan (2003) (Tsai *et al*., 2003), China (Wu *et al*., 2003), Serbia (2005) (Serbulaa *et al*., 2010), Serbia (2006) (Serbulaa *et al*., 2010), Serbia (2007) (Serbulaa *et al*., 2010), Greece (2008) (Tsopelas *et al*., 2008) and China (2009) (Lu *et al*., 2009), respectively. From the point of view of $PM_{2.5}$, the results indicated that the maximum average particulate concentration for As was occurred at Spanish (2002) (average, 7.9 ng/m^3) (Sa'nchez de la Campa *et al*., 2008). This value was about 1.2, 2.1, 10, 2.6, 1.6 and 1.6 times higher than those of Spanish (2001) (Sa'nchez de la Campa *et al*., 2008), Taiwan (2003) (Tsai *et al*., 2003), Greece (2003) (Thomaidis *et al*., 2003), Spain (2004) (Fernández-Camacho *et al*., 2010), Hong Kong (2005) (Louie *et al*., 2005), and Hong Kong (2007) (Hagler *et al*., 2007), respectively. Finally from the point of view of TSP, the results indicated that the maximum average particulate concentrations for As were occurred at Spain (2000) (average, 12.3 ng/m^3) (Oliveira *et al*., 2005). This value was about 3.1, 3.2, 3.7, 2.9, and 3.9 times as those of Bei-shi, Chang-hua, He-mei, Quan-xing and Gao-mei, respectively.

Table 6. Comparison of ambient air As concentrations (ng/m^3) at various particle sizes for different characteristic sampling sites during year of 2000–2010

Year	Reference	Location	Characteristic	Size	As (ng/m^3)
2000	Oliveira *et al*. (2005)	Spain	industrial	TSP	12.3
2001	Sanchez-Rodas *et al*. (2007)	Huelva	urban	PM_{10}	7.7
	Sa'nchez de la Campa *et al*. (2008)	Spanish	urban	$PM_{2.5}$	6.4
2002	Sanchez-Rodas *et al*. (2007)	Huelva	urban	PM_{10}	9.9
	Sa'nchez de la Campa *et al*. (2008)	Spanish	urban	$PM_{2.5}$	7.9
2003	Tsai *et al*. (2003)	Taiwan	urban	PM_{10}	6.0
	Tsai *et al*. (2003)			$PM_{2.5}$	3.8
	Wu *et al*. (2003)	China	urban	PM_{10}	17.0
	Thomaidis *et al*. (2003)	Greece	industries	$PM_{2.5}$	0.8
2004	Zheng *et al*. (2004)	China	urban	PM_{10}	42.1
	Fernández-Camacho *et al*. (2010)	Spain	University	$PM_{2.5}$	3.0
2005	Serbulaa *et al*. (2010)	Serbia	urban	PM_{10}	29.3
	Louie *et al*. (2005)	Hong Kong	urban	$PM_{2.5}$	5.0
2006	Serbulaa *et al*. (2010)	Serbia	urban	PM_{10}	38.9
2007	Serbulaa *et al*. (2010)	Serbia	urban	PM_{10}	25.0
	Hagler *et al*. (2007)	Hong Kong	urban	$PM_{2.5}$	4.8
2008	Tsopelas *et al*. (2008)	Greece	industrial	PM_{10}	1.1
2009	Lu *et al*. (2009)	China	industries	PM_{10}	6.2
2009–2010	This study	Taiwan	suburban/coastal	TSP	3.30
2009–2010	This study	Taiwan	downtown	TSP	3.32
2009–2010	This study	Taiwan	residential	TSP	2.92
2009–2010	This study	Taiwan	industrial	TSP	3.64
2009–2010	This study	Taiwan	wetland	TSP	2.83

Fig. 2 displays the calculated/measured flux ratio for the atmospheric pollutant arsenic (As) at Bei-shi (suburban/ coastal) sampling site. The results demonstrate that the calculated/measured flux ratios of arsenic (As) were from 0.571 to 1.747. The average calculated/measured flux ratio was 1.013.

Fig. 3 displays the calculated/measured flux ratio for atmospheric pollutant arsenic (As) at Chang-hua (downtown) sampling site. The range of calculated/measured flux ratios was from 0.883 to 1.018. The average was 1.001.

Fig. 4 displays the calculated/measured flux ratio at the He-mei (residential) sampling site. The range of the calculated/measured flux ratio was from 0.549 to 1.251, and the average was 0.999.

Fig. 5 displays the calculated/measured flux ratio for atmospheric pollutant arsenic (As) at the Quan-xing (industrial) sampling site. The range of calculated/ measured flux ratios was from 0.641 to 2.096, and the average was 1.029.

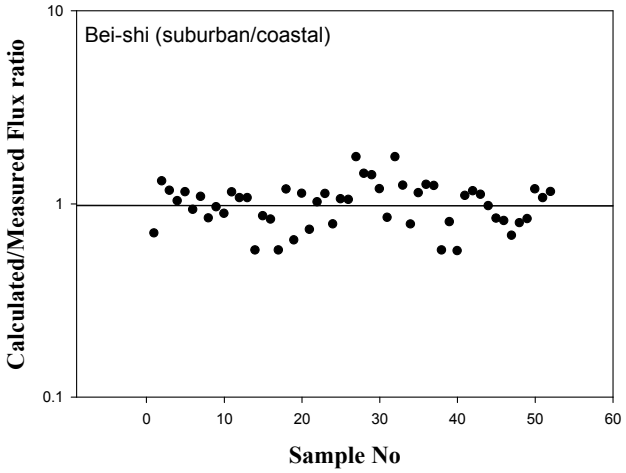

Fig. 2. Calculated/Measured flux ratio for atmospheric pollutant arsenic (As) at Bei-shi (suburban/coastal) sampling site.

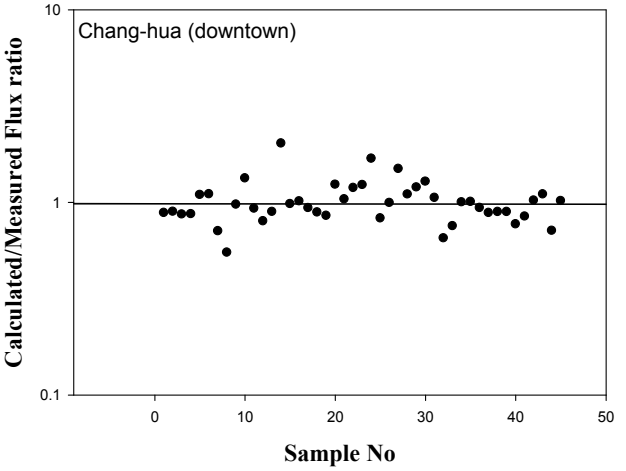

Fig. 3. Calculated/Measured flux ratio for atmospheric pollutant arsenic (As) at Chang-hua (downtown) sampling site.

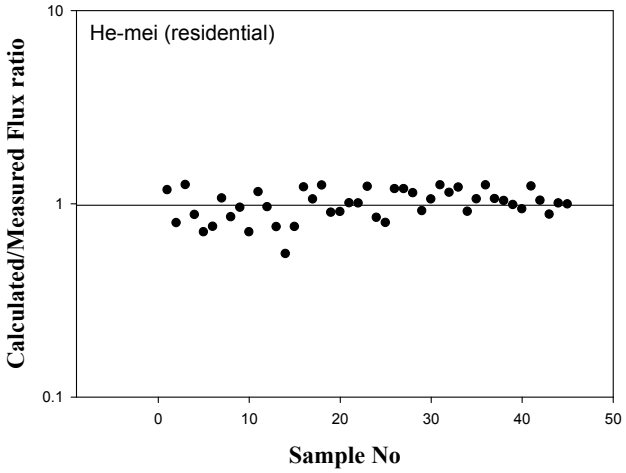

Fig. 4. Calculated/Measured flux ratio for atmospheric pollutant arsenic (As) at He-mei (residential) sampling site.

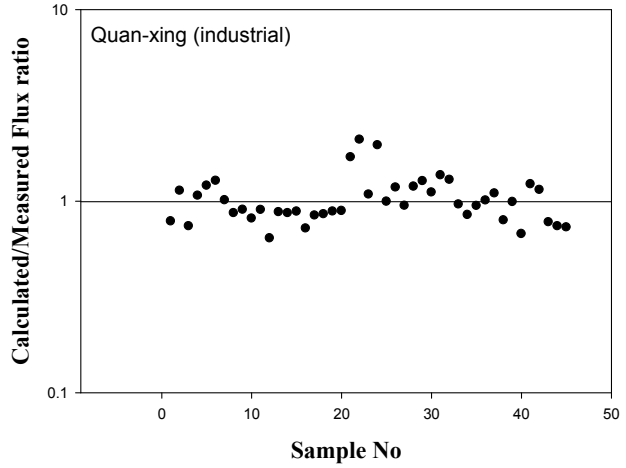

Fig. 5. Calculated/Measured flux ratio for atmospheric pollutant arsenic (As) at Quan-xing (industrial) sampling site.

Fig. 6 plots the calculated/measured flux ratio for atmospheric pollutant arsenic (As) at the Gao-mei (wetland) sampling site. The range of the calculated/measured flux ratio for arsenic (As) was from 0.618 to 1.361, and the average was 1.007.

The calculated/measured flux ratio was fallen into an appropriate ratio which indicated that this model can apply to in the prediction of the dry deposition for these sampling sites.

CONCLUSIONS

1. The fluxes of dry-deposited arsenic (As) followed the order, Quan-xing (industrial) > He-mei (residential) > Bei-shi (suburban/coastal) > Chang-hua (downtown) > Gao-mei (wetland). The seasonal variations of As dry-deposited were in the order fall > winter > spring > summer at He-mei (residential) and Gao-mei (wetland), respectively.

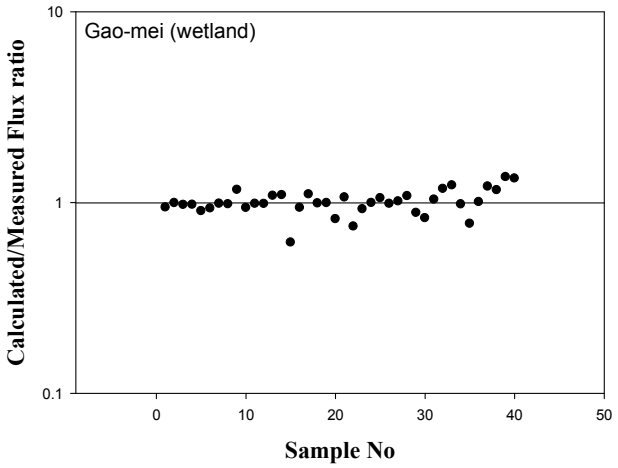

Fig. 6. Calculated/Measured flux ratio for atmospheric pollutant arsenic (As) at Gao-mei (wetland) sampling site.

2. The arsenic (As) concentrations in total suspended particulates (TSP) followed the order Quan-xing (industrial) > Chang-hua (downtown) > Bei-shi (suburban/coastal) > He-mei (residential) > Gao-mei (wetland). The seasonal variations of As concentrations in TSP were in the order fall > winter > spring > summer at Bei-shi (suburban/coastal), Quan-xing (industrial) and Gao-mei (wetland), respectively.
3. The arsenic (As) compositions in TSP followed the order He-mei (residential) > Quan-xing (industrial) > Bei-shi (suburban/coastal) > Chang-hua (downtown) > Gao-mei (wetland). The seasonal variations of As compositions in TSP were in the order winter > fall > spring > summer at He-mei (residential) and Quan-xing (industrial), respectively.
4. The best-fit overall dry deposition velocities exhibited excellently predicted ambient air arsenic (As) dry deposition fluxes. The average calculated/measured flux ratios for As at Chang-hua (downtown), He-mei (residential) and Gao-mei (wetland) were 1.001, 0.999 and 1.007, respectively. However, the mean calculated/measured flux ratios for As at Bei-shi (suburban/coastal) and Quan-xing (industrial) were 1.013 and 1.029, respectively.
5. The calculated/measured flux ratio was fallen into an appropriate ratio which indicated that this model can apply to in the prediction of the dry deposition for these sampling sites.

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