Measuring and Modeling Atmospheric Arsenic Pollutants, Total As, As(III), and As(V), at Five Characteristic Sampling Sites

Guor-Cheng Fang[*], Jun-Han Huang, Chia-Kuan Liu, Yi-Liang Huang

Department of Safety, Health and Environmental Engineering, HungKuang University, Sha-Lu, Taichung 433, Taiwan

ABSTRACT

This investigation determines ambient air arsenic (As), As(III), and As(V) concentrations in total suspended particulates (TSP) and dry deposition. Calculated/measured dry deposition flux ratios of ambient air As, As(III) and As(V) were evaluated using two dry deposition models at five characteristic sampling sites during 2009–2010.The highest average concentrations of As, As(III) and As(V) in TSP and dry deposition were measured at the Quan-xing industrial sampling site during August–January. The Quan-xing site, with many industrial factories under process around there regions, its air is extremely polluted. In addition, the average dry deposition velocities for ambient air total arsenic (As) at Bei-shi, Chang-hua, He-mei, Quan-xing and Gao-mei were 0.60 (cm/sec), 0.57 (cm/sec), 0.73 (cm/sec), 0.67 (cm/sec) and 0.66 (cm/sec), respectively at these five characteristic sampling sites.

The highest average seasonal variation for As, As(III) and As(V) in TSP and dry deposition were in winter and fall due to emissions from fossil fuel combustion by the nearby Taichung Thermal Power Plant (TTPP) and for household heating. The Noll and Fang model can be applied to predict dry deposition of As, As(III), and As(V) at these sampling sites.

Keywords: TSP; Dry deposition; Model; Arsenic (As); As(III); As(V).

INTRODUCTION

Atmospheric particulate matter (PM) in urban areas has many sources, the majority of which are closely related to human activity. Heavy metals emissions from factories or car exhausts can result in serious environmental problems such as the restriction of atmospheric visibility, while their toxicity may present health problems to humans at certain concentrations (Milford and Davidson, 1985). Principal sources of 13 metallic elements in PM included natural sources and local anthropogenic sources such as non-ferrous metal smelting, oil combustion, welding, vehicular traffic and road dust (Ny and Lee, 2011). The that point-source emissions were the predominant contributors (about 49.1%) to PM_{10} concentrations at Hsiung-Kong site industrial site in Kaohsiung City, followed by area sources (approximately 35.0%) and transport from neighboring areas (7.8%) (Wang and Chen, 2008). Results showed that major contribution of $PM_{2.5}$ were the mobile source emissions with 45% (Vega *et al.*, 2010). And then, in the industrial emissions of the target pollutants depended upon the subareas and chemical types (Choi and Jo, 2011).

The predominance of a particular wind direction determines the distribution of PM and As (Serbula *et al.*, 2010). Road transport was considered the dominant source of PM_{10} (Inoka Senaratne *et al.*, 2005). Therefore, traffic related nano and ultrafine particles are possibly cytotoxic (Lin *et al.*, 2008). The results show that RDS0.5 has significantly lower particulate matter (PM) emissions, while the PM emissions of PFO 0.5 are higher than those of FBFA (Wu *et al.*, 2010).

However, motor vehicles containing organic and elemental components that are typically found in fine particles were included in coarse particle mass during dust events (Stone *et al.*, 2011). Fine particles emitted from vehicles have adverse health effects because of their sizes and chemical compositions (Lin *et al.*, 2005). The concentration levels of particulate matter (PM) in the city of Beijing (39.92°N, 116.46°E), China are dependent on the long-range transport of PM in addition to local stationary and mobile sources (Xu *et al.*, 2008). 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).

Accordingly, high dry deposition velocities are threatening to human health by dry deposition (Chang *et al.*, 2003). The experimental results show that PM_{10} concentration in the fall was higher than in spring and summer (Tsai *et al* 2010). High concentrations were recorded in the winter seasons for both particulate pollutants. In winter season,

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[*] Corresponding author. Tel: (886)-4-26318652 ext. 1110;
  Fax: (886)-4-2631-0744
  E-mail address: gcfang@sunrise.hk.edu.tw

temperature is low and wind speed is generally low (Vijay Bhaskar and Mehta, 2010). These particulate pollutants concentrations were compared with meteorological variables such as rainfall, humidity, temperature, and wind speed. Both SPM and PM_{10} showed significant negative correlations with rainfall (Vijay Bhaskar and Mehta, 2010).The differences in the results at the two sites are mainly attributed to the different industries at each site. The mean deposition fluxes of airborne particles were between 182.2 and 195.3 mg/m^2/day, with a dry deposition velocity of 2.00–2.04 cm/s (Wang *et al.*, 2007).

Seasonal differences in the metal concentrations may be due to differences in wind directions with some winds passing through industry or traffic areas (Lee and Hieu, 2011). Results indicate that summer has a significantly higher concentration of the metals Zn, Pb, Cu, Cd than winter and monsoon (Saxena *et al.*, 2008). Determination of metals composition of inhalable particles is important in determining their potential impact on human health (Park *et al.*, 2008).

Arsenic, present in both organic and inorganic forms in natural environment, is toxic to humans and elements for human health that persist and cycle in the environment as a result of natural and anthropogenic activities (Soros *et al.*, 2003; Walcek *et al.*, 2003). Arsenic is considered to be one of the most toxic elements for human health. Continued exposure to a high concentration of arsenic may produce an acute toxic effect on humans, which can be quickly diagnosed. However, low doses of arsenic do not result in an acute toxic effect, but may give rise to cancer after prolonged exposure (Hayes, 1997; Roy and Saha, 2002).

The study measures atmospheric particulates arsenic (As), As(III) and As(V) concentration in total suspended particulates (TSP) and the dry deposition of As, As(III), and As(V) seasonally. This study also compares calculated/ measured dry deposition flux ratios of ambient air As, As(III) and As(V) at five characteristic sampling sites- suburban/coastal, downtown, residential, industrial, and wetland sites- with those calculated using two dry deposition models.

METHODOLOGY

Sampling Program

Fig. 1 lists the five characteristic sampling sites. All samples were obtained in 1380–1400 min during the sampling period for each sampling group. The sampling sites were as follows:

The Bei-shi sampling station (24°13'31.82"N, 120°34'09.45"E), which is in Shalu, Taichung, Taiwan, is a suburban/coastal station with no nearby obstructions. The immediate vicinity is residential, with an expressway with heavy traffic located approximately 2 km east of the station. The Chang-Hua sampling station (24°05'24.52"N, 120°31'31.73"E) is within the city, high levels of vehicle emissions, and emissions from a chemical plant within the city. The He-mei town sampling station (24°06'00.54"N, 120°30'51.34"E) is located in a residential area. The main pollution sources are resident activities and vehicular emissions. The Quan-xing sampling station (24°08'37.89"N, 120°29'09.43"E) is located in Shen-kang, a town covering 246.8 hectares, roughly half of which, 126.5 hectares, is occupied by factories and industry. The fifth sampling station is the Gao-Mei wetland station (24°18'35.07"N,
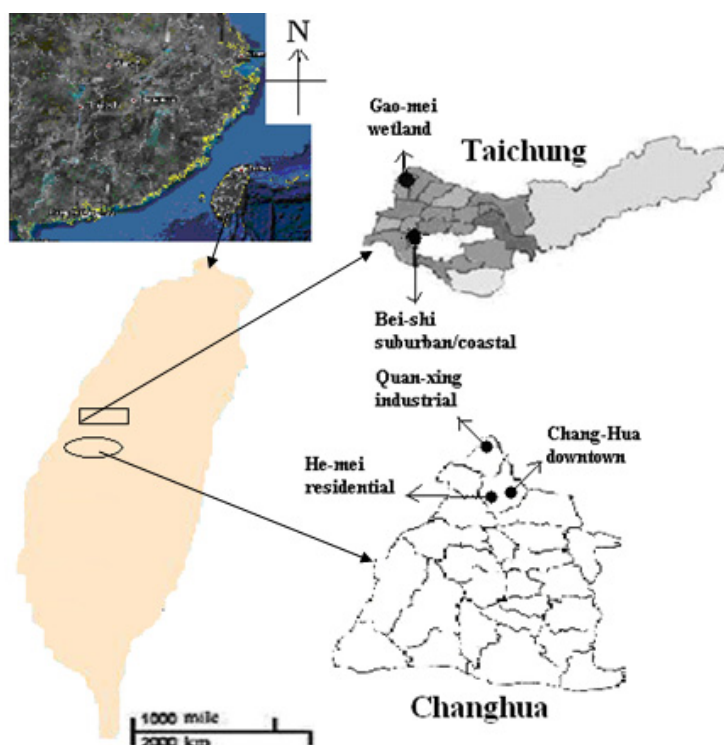

Fig. 1. Geographical location at five characteristic sampling sites in central Taiwan.

120°33'08.21"E), which is located in Shimizu, Taichung, Taiwan. This wetland covers over 300 hectares and is located in central Taiwan. The wetland is home to seven habitats: tidal creek area, swamp area, sand area, gravel area, and Yunlin Wan grass areas. Additionally, the Taichung Thermal Power Plant (TTPP) sits on 281 hectares of the wetland along the coast, on the western side of the sampling site. This plant burns coal to supply central Taiwan with 4,400 MW of electricity daily.

Sampling Program

PS-1 Sampler

The PS-1 is a complete air sampling system designed to collect suspended airborne particles (GMW High-Volume Air Sampler; Graseby-Andersen, country). Maximum particle size in this study was roughly 100 μm. Sampler flow rate was 200 L/min. A quartz filter 10.2 cm in diameter filtered suspended particles. Filters were first conditioned for 24 h under an electric chamber under a humidity of humidity 35 ± 5% and temperature 25 ± 5°C prior to both on and off weighing. Filters were in a sealed CD box when transported and stored. The sampling device and procedures are similar to those in previous study (Fang *et al.*, 2010).

Dry Deposition plate

The dry deposition plate (DDP) had a smooth horizontal surrogate surface, providing a lower-bound estimate of dry deposition flux. The polyvinyl chloride (PVC) DDP measured 21.5 cm long, 8.0 cm wide by 0.8 cm thick. The DDP also had a sharp leading edge that was pointed into the prevailing wind. All filters were maintained under 50% relative humidity at 25°C for over 48 h. Prior to sampling, all filters were weighed to 0.0001 Gram-significant digits (Fang *et al.*, 2009).

Chemical Analysis

The samples were placed in an oven one night before being weighed. A quarter of the filter was cut and selected before the digestion process. The filtered were cut into pieces thin added into the Teflon cup. 3 mL of hydrochloric acid (HCl) and 9 mL of nitrate (HNO_3) were mixed and then added to this cup. After that, the samples were heated at 500C on the hot plate for two hours. Samples after digestion on the hot plate will then be filtered. After filtration, the sample solution will then be added 0.2% of HNO_3 and added up to 100 mL. solution. Before ICP-AES analysis, there samples will be kept 40C in the refrigerator.

Quality Control

Blank test background contamination was assessed using operational blanks (unexposed projection film and a quartz filter) that 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 filters. This study accounted for background contamination of As by subtracting field blank values from concentrations. Field blank values were extremely low, generally below or around the method detection limits. In this study, the background contamination is insignificant and can be ignored. Blank

test results were 0.19, 0.05, and 0.10 μg for As, As(III) and As(V), respectively.

Dry Deposition Models

Atmospheric particles with aerodynamic diameters < 10 μm (PM_{10}) have been under scrutiny as they are easily inhaled and deposited within the respiratory system (Pope *et al.*, 1995). Studies show that PM_{10} has a role in the incidence and severity of respiratory diseases (Pope and Dockery, 1999; Brunekreef and Holgate, 2002). Therefore, 10 μm was used to calculate Vd, and modeled dry deposition for comparison with measured dry deposition fluxes at the five characteristic sampling sites. In addition, best-fit overall method has been successfully applied in the modeling and estimating of ambient air arsenic pollutant in previous study (Fang *et al.*, 2011). This study further applied two different dry deposition model in the modeling and estimating ambient air arsenic pollutants, total As, As(III), and As(V) at five characteristic sampling sites.

The following two dry deposition models were applied herein study.

Baklanov and Sorensen's Model

Baklanov and Sorensen (2001) developed deposition models for computing long-range deposition. They defined dry deposition velocity as the inverse of the sum of resistances r_a, r_b, and r_c in three sequential layers (Fang *et al.*, 2006; Basu *et al.*, 2009). The model takes the following form for gaseous pollutants.

$$V_d = (r_a + r_b + r_c)^{-1}, \tag{1}$$

where r_a denotes the aerodynamic resistance; r_b is the resistance to penetration across the atmospheric laminar sublayer, and r_c is the resistance associated with direct pollutant-surface interaction.

Baklanov and Sorensen, 2001 suggested that transfer resistance, r_c, is negligible for particles, since once a particle encounters a surface, it is considered to be deposited. For particles, Seinfield suggested the use of the term, $r_a r_b v_g$, rather than r_c.

The above formula for the dry deposition velocity of particles has a term that is determined by the sedimentation/ gravitational settling of particles:

$$V_d = (r_a + r_b + r_a r_b v_g)^{-1} + v_g, \tag{2}$$

where v_g is the gravitational settling velocity.

The aerodynamic resistance r_a depends on meteorological parameters, such as wind speed, atmospheric stability and surface roughness, as follows.

$$r_a = [\ln(Z_s/Z_o) - \Psi_c]/k u_*, \tag{3}$$

Surface layer resistance, r_b, depends on parameters of diffusion across a laminar sublayer. Therefore, this resistance depends on molecular rather than turbulent characteristics. Therefore, surface layer resistance for particles differs from that for gases. According to Zannetti (1990), surface layer

resistance for particles can be expressed as a function of the Schmidt number, Sc = v/D, and Stokes number, St:

$$r_b = (S_c^{-2/3} + 10^{-3/St})^{-1} u_*^{-1} \qquad (4)$$

For particles with diameters < 3.5 μm, airflow around a falling particle can be considered laminar However, for particles with larger diameters, Stokes law does not hold, and an iterative procedure must be be used to solve the equation for terminal settling velocity in the turbulent regime (Näslund and Thaning, 1991):

$$Dw_p/dt = (w - w_p)f(V) - \beta g, \qquad (5)$$

$$f(V) = 3\rho V C_d/8r\rho_p, \qquad (6)$$

$$V = ((u - u_p)^2 + (v - v_p)^2 + (w - w_p)^2)^{1/2}, \qquad (7)$$

$$C_d = 24/Re[1 + 0.173(Re)^{0.657}] + 0.413/(1 + 16300(Re)^{-1.09}); \qquad (8)$$

where V is the relative velocity of particles u, v, w, u_p, v_p and w_p are the air and particle velocity components, β is the buoyancy effect parameter, $\beta = (\rho_p - \rho)/\rho_p$, C_d is the drag coefficient in the static state, and Re is the Reynolds number, Re = 2Vr/v.

Noll & Fang's Model

Noll & Fang's dry deposition model yield the following deposition velocities of atmospheric particles (Fangh, 1989).

$$V_d = V_{st} + 1.12U^* \chi \exp(-30.36/D_p) \qquad (9)$$

where, V_d = particle settling velocity (cm/s); U^* = (friction velocity : *clear to a technical reader?*) (cm/s); and D_p = particle diameter (μm).

Two meteorological parameters that influence atmospheric turbulence are frictional velocity U^* and surface roughness Z_0. The relationship between these parameters for near nature atmospheric stability conditions is

$$U = (U^*/\kappa)* \ln [(Z-d)/ Z_0] \qquad (10)$$

where, U = measured average wind speed at height Z (m/s); Z = measured height above ground (m); k = Von Karmon's constant (0.4); d = displacement (m), and Z_0 = surface roughness height (m).

RESULTS AND DISCUSSION

Table 1 displays shows the meteorological conditions and arsenic (As), As(III) and As(V) concentrations in total suspended particulates (TSP) and dry deposition at the five sampling sites during year 2009–2010. Mean temperature was 24–25°C; average wind speed was 1.97–2.69 m/sec; the average relative humidity was 72–79% at the five sampling sites.

The highest average concentration of As in TSP at the Quan-xing industrial site was 3.55 ng/m³ and the lowest

average concentration in TSP at the He-mei residential site was 2.79 ng/m³. The ratios for these two values were about 1.27. At the Quan-xing industrial site, the highest average concentration of As(III) in TSP was 1.06 ng/m³ and the lowest average concentration in TSP at the He-mei residential site was 0.83 ng/m³. The ratios for these two values were about 1.28. The highest average As(V) concentration in TSP at the Quan-xing industrial site was 2.48 ng/m³ and the lowest average concentration in TSP at the He-mei residential site was 1.94 ng/m³. The ratios for these two values were about 1.28. Moreover, the highest average seasonal concentrations for As, As(III) and As(V) in TSP occurred during fall and lowest average seasonal concentrations occurred during summer at the Bei-shi suburban/coastal, Quan-xing industrial, and Gao-mei wetland sampling sites. Additionally, the highest average seasonal concentration for As, As(III) and As(V) in TSP occurred during winter and the lowest average seasonal concentration in TSP occurred during summer at the Chang-hua downtown sampling site. Finally, the highest average seasonal concentration for As, As(III), and As(V) in TSP occurred during spring and the lowest average seasonal concentration in TSP was in summer at the He-mei residential sampling site.

The highest average of As dry deposition at the Quan-xing industrial site was 2.36 ng/m²/min and lowest average dry deposition at the Gao-mei wetland site was 1.80 ng/m²/min. The ratios for these two values were about 1.31. Additionally, analytical results indicate that the highest average As(III) dry deposition at the Quan-xing industrial was 0.71 ng/m²/min and lowest average dry deposition at the Gao-mei wetland site was 0.54 ng/m²/min. The ratios for these two values were about 1.31. And the highest average of As(V) dry deposition at the Quan-xing industrial site was 1.67 ng/m²/min and lowest average dry deposition at the Gao-mei wetland site was 1.26 ng/m³. The ratios for these two values were about 1.33. In addition, the results indicated that the highest average dry deposition velocity for ambient air total arsenic (As) at the He-mei was 0.76 cm/sec. Additionally, the results indicated that the lowest average dry deposition velocity for ambient air total arsenic (As) at the Chang-hua was 0.57 cm/sec. Moreover, the highest average seasonal dry deposition for As, As(III) and As(V) were in fall and the lowest average seasonal dry deposition were in summer at the Chang-hua downtown site, He-mei residential site and Gao-mei wetland site. Additionally, the highest average seasonal dry deposition for As, As(III) and As(V) were in winter and the lowest average seasonal dry deposition were in summer at the Bei-shi suburban/coastal site. Finally, the highest average seasonal dry deposition for As, As(III) and As(V) were in fall and the lowest average seasonal dry deposition were in spring at the Quan-xing industrial site.

The main As, As(III), and As(V) sources at the He-mei residential sampling site were fossil fuel combustion for transportation, heating, and waste incineration. At the Chang-hua downtown sampling site, nearby sources of As, As(III), and As(V) were transportation, chemical plant and fossil fuel combustion. The main sources of As, As(III),

Table 1. Meteorological conditions and arsenic (As), As (III) and As (V) in total suspended particulates (TSP), dry deposition and dry deposition velocity at five characteristic sampling sites during year 2009–2010.

Sites	Sample date	Meteorological conditions			TSP (ng/m3)			Dry deposition (ng/m2/min)			Vd (cm/sec)
		Temp (°C)	WS (m/sec)	RH (%)	As	As(III)	As(VI)	As	As(III)	As(VI)	As
Bei-shi											
	Feb./'10	18.67	1.55	87.5	3.42	1.03	2.39	1.53	0.46	1.09	0.45
Spring	Mar./'10	22.78	1.72	84.1	2.97	0.89	2.08	1.54	0.46	1.1	0.52
	Apr./'10	22.38	2.03	79.55	3.21	0.96	2.24	2.14	0.64	1.51	0.67
	Mean	21.27	1.77	83.72	3.2	0.96	2.24	1.74	0.52	1.23	0.54
	May/'10	27.63	2.29	77.51	2.9	0.87	2.03	1.83	0.55	1.27	0.63
Summer	Jun./'10	27.15	2.66	87.49	2.62	0.79	1.82	1.71	0.51	1.2	0.65
	Jul./'10	27.9	3.17	84.62	1.99	0.6	1.36	1.37	0.41	0.99	0.69
	Mean	27.56	2.71	83.21	2.5	0.75	1.74	1.64	0.49	1.15	0.65
	Aug./'10	29.6	2.29	74.94	3.43	1.03	2.38	1.93	0.58	1.31	0.56
Fall	Sep./'09	28.91	1.89	70.32	3.47	1.04	2.46	1.95	0.58	1.42	0.56
	Oct./'09	24.56	1.48	65.99	3.61	1.08	2.53	2.12	0.64	1.5	0.59
	Mean	27.69	1.89	70.42	3.5	1.05	2.46	2	0.6	1.41	0.57
	Nov./'09	22.91	1.44	76.11	3.37	1.01	2.38	2.23	0.67	1.58	0.66
Winter	Dec./'09	19.39	1.61	70.27	3.18	0.96	2.27	2.22	0.67	1.54	0.70
	Jan./'10	17.39	1.1	69.84	3.6	1.08	2.5	1.88	0.56	1.33	0.52
	Mean	19.89	1.38	72.08	3.39	1.02	2.38	2.11	0.63	1.48	0.62
Average		24.28	1.97	77.46	3.13	0.94	2.19	1.87	0.56	1.32	0.60
Chang-hua											
	Feb./'10	17.85	1.39	78.13	3.91	1.17	2.7	1.79	0.54	1.3	0.46
Spring	Mar./'10	24.2	1.63	74.62	3.09	0.93	2.21	1.92	0.58	1.36	0.62
	Apr./'10	20.27	1.81	71.31	3.31	0.96	2.37	2.07	0.6	1.47	0.63
	Mean	20.77	1.61	74.69	3.44	1.02	2.43	1.93	0.57	1.38	0.56
	May/'10	28.66	2.59	68.23	2.81	0.84	2.01	1.67	0.5	1.18	0.59
Summer	Jun./'10	27.27	2.38	81.41	2.5	0.74	1.76	1.48	0.44	1.13	0.59
	Jul./'10	28.1	2.92	77.73	2.22	0.67	1.53	1.3	0.39	0.89	0.58
	Mean	28.01	2.63	75.79	2.51	0.75	1.77	1.48	0.44	1.07	0.59
	Aug./'10	30.02	2.39	74.97	3.28	0.98	2.3	1.95	0.59	1.37	0.60
Fall	Sep./'09	30.5	1.57	66.83	3.32	1	2.3	1.98	0.59	1.4	0.60
	Oct./'09	26.22	1.02	69.62	3.23	0.97	2.33	2	0.6	1.43	0.62
	Mean	28.91	1.66	70.47	3.28	0.98	2.31	1.98	0.59	1.4	0.60
	Nov./'09	25.02	1.26	70.66	3.34	1	2.34	1.79	0.54	1.22	0.54
Winter	Dec./'09	19.55	2.33	69.08	3.15	1.15	2.68	2.13	0.65	1.5	0.68
	Jan./'10	19.12	1.23	68.21	3.68	1.11	2.54	1.76	0.53	1.27	0.48
	Mean	21.23	1.61	69.32	3.39	1.09	2.52	1.89	0.57	1.33	0.56
Average		24.89	1.84	72.67	3.19	0.95	2.24	1.81	0.54	1.29	0.57
He-mei											
	Feb./'10	18.02	2.24	81.3	2.95	0.88	2.05	1.92	0.58	1.35	0.65
Spring	Mar./'10	23.91	1.72	78.49	2.96	0.89	2.07	1.9	0.58	1.35	0.64
	Apr./'10	20.33	3.21	73.79	3.04	0.86	2.1	2.05	0.59	1.45	0.68
	Mean	20.75	2.39	77.86	2.98	0.88	2.07	1.96	0.58	1.38	0.66
	May/'10	28.14	2.75	73.79	2.74	0.82	1.91	1.91	0.58	1.36	0.70
Summer	Jun./'10	26.96	2.52	84.96	2.4	0.7	1.69	1.67	0.5	1.2	0.70
	Jul./'10	27.82	3.09	79.91	2.09	0.63	1.47	1.5	0.45	1.06	0.72
	Mean	27.64	2.79	79.56	2.41	0.72	1.69	1.7	0.51	1.2	0.70
	Aug./'10	29.52	2.49	77.23	2.89	0.86	2.01	2.19	0.66	1.54	0.76
Fall	Sep./'09	30.44	1.88	69.02	2.96	0.88	2.05	2.23	0.68	1.58	0.76
	Oct./'09	26.16	1.83	71.36	2.86	0.86	2.02	2.37	0.71	1.65	0.83
	Mean	28.7	2.07	72.54	2.9	0.87	2.02	2.26	0.68	1.59	0.78
	Nov./'09	24.9	1.1	73.77	2.6	0.77	1.79	2.29	0.7	1.63	0.88
Winter	Dec./'09	19.55	2.33	69.08	3.15	0.94	2.2	2.13	0.65	1.51	0.68
	Jan./'10	19.06	1.73	71.93	3.03	0.91	2.12	2.22	0.67	1.56	0.73

Table 1. *(continued).*

Sites	Sample date	Meteorological conditions			TSP (ng/m^3)			Dry deposition (ng/m^2/min)			Vd (cm/sec)
		Temp (°C)	WS (m/sec)	RH (%)	As	As(III)	As(VI)	As	As(III)	As(VI)	As
	Mean	21.17	1.72	71.59	2.93	0.87	2.04	2.21	0.67	1.57	0.76
Average		24.74	2.26	75.64	2.79	0.83	1.94	2.02	0.61	1.43	0.73
Quan-xing											
Spring	Feb./'10	18.19	3.1	84.47	3.67	1.1	2.56	2.07	0.62	1.45	0.56
	Mar./'10	23.62	1.82	82.37	3.46	1.02	2.38	2.06	0.62	1.45	0.60
	Apr./'10	20.38	4.6	76.26	3.48	1.03	2.4	2.47	0.71	1.75	0.71
	Mean	20.73	3.17	81.03	3.53	1.05	2.45	2.2	0.65	1.55	0.62
Summer	May/'10	27.62	2.91	79.36	3.2	0.94	2.19	2.27	0.69	1.6	0.71
	Jun./'10	26.66	2.67	88.52	3.15	0.93	2.17	2.24	0.66	1.59	0.71
	Jul./'10	27.54	3.27	82.09	3.01	0.89	2.08	2.25	0.68	1.59	0.75
	Mean	27.27	2.95	83.32	3.12	0.92	2.15	2.25	0.68	1.6	0.72
Fall	Aug./'10	29.02	2.59	79.49	3.81	1.16	2.7	2.51	0.75	1.75	0.66
	Sep./'09	30.37	2.19	71.2	3.85	1.17	2.74	2.53	0.76	1.76	0.66
	Oct./'09	26.1	2.64	73.1	3.88	1.16	2.71	2.61	0.8	1.87	0.67
	Mean	28.49	2.47	74.6	3.85	1.16	2.71	2.55	0.77	1.79	0.66
Winter	Nov./'09	24.78	0.93	76.88	3.74	1.1	2.58	2.89	0.86	2.01	0.77
	Dec./'09	19.84	3.25	71.05	3.6	1.1	2.57	2.76	0.83	1.94	0.77
	Jan./'10	18.99	2.23	75.65	3.93	1.17	2.74	1.74	0.53	1.23	0.44
	Mean	21.21	2.14	74.53	3.76	1.13	2.63	2.46	0.74	1.73	0.66
Average		24.58	2.69	78.61	3.55	1.06	2.48	2.36	0.71	1.67	0.67
Gao-mei											
Spring	Feb./'10	18.48	2.35	82.56	2.72	0.81	1.89	1.86	0.56	1.3	0.68
	Mar./'10	23.95	1.66	80.82	2.69	0.81	1.89	1.84	0.55	1.29	0.68
	Apr./'10	20.68	3.52	75.49	2.99	0.9	2.1	1.92	0.58	1.35	0.64
	Mean	21.04	2.51	79.62	2.8	0.84	1.96	1.87	0.56	1.31	0.67
Summer	May/'10	27.84	2.6	77.34	2.66	0.78	1.9	1.44	0.41	1.01	0.54
	Jun./'10	27.51	2.34	81.99	2.5	0.75	1.76	1.36	0.4	0.94	0.54
	Jul./'10	28.44	2.85	78.59	2.37	0.7	1.67	1.24	0.37	0.88	0.53
	Mean	27.93	2.6	79.31	2.51	0.74	1.78	1.35	0.39	0.94	0.54
Fall	Aug./'10	29.31	2.12	76.12	3.03	0.91	2.13	2.08	0.62	1.46	0.69
	Sep./'10	27.86	1.96	77.41	3.07	0.92	2.16	2.1	0.63	1.47	0.68
	Oct./'09	26.12	1.89	73.07	3.11	0.93	2.18	2.13	0.64	1.49	0.69
	Mean	27.77	1.99	75.53	3.07	0.92	2.16	2.1	0.63	1.47	0.69
Winter	Nov./'09	24.66	0.99	76.79	3.03	0.91	2.13	2	0.6	1.4	0.66
	Dec./'09	19.85	2.46	70.55	2.72	0.81	1.89	1.89	0.57	1.32	0.70
	Jan./'10	19.03	1.83	75.08	2.68	0.8	1.86	1.83	0.54	1.27	0.68
	Mean	21.18	1.76	74.14	2.81	0.84	1.96	1.91	0.57	1.33	0.68
Average		24.72	2.22	77.24	2.8	0.84	1.96	1.8	0.54	1.26	0.64

and As(V) at the Bei-shi suburban/coastal sampling site were the science park, fossil fuel combustion and transportation. At the Gao-mei wetland sampling site, the sources of As, As(III), and As(V) were the TTPP and fossil fuel combustion. Finally, the main sources of As, As(III), and As(V) at the Quan-xing industrial sampling site were steel industry, electronic industry, plastic industry, chemical industry, basic metal manufacturing, machinery manufacturing, and petroleum and coal products.

Fig. 2 shows the calculated/measured flux ratios for As at the five sampling sites (A) Bei-shi (suburban/coastal) (B) Chang-hua (downtown) (C) He-mei (residential) (D) Quan-xing (industrial) (E) Gao-mei (wetland) the two deposition

models. The average calculated/measured As flux ratios by the Baklanov model at the Bei-shi, Chang-hua, He-mei, Quan-xing, and Gao-mei sites were 7.56, 7.91, 6.16, 6.85, and 6.98, respectively. Finally, average calculated/measured flux ratios for As by Noll and Fang model at the Bei-shi, Chang-hua, He-mei, Quan-xing, and Gao-mei sites were 1.60, 1.68, 1.31, 1.45 and 1.48, respectively.

Fig. 3 plots calculated/measured flux ratios for As(III) at the five sampling sites (A) Bei-shi (suburban/coastal) (B) Chang-hua (downtown) (C) He-mei (residential) (D) Quan-xing (industrial) (E) Gao-mei (wetland) the two deposition models. The average calculated/measured flux ratios by the Baklanov model for As(III) at the Bei-shi, Chang-hua, He-
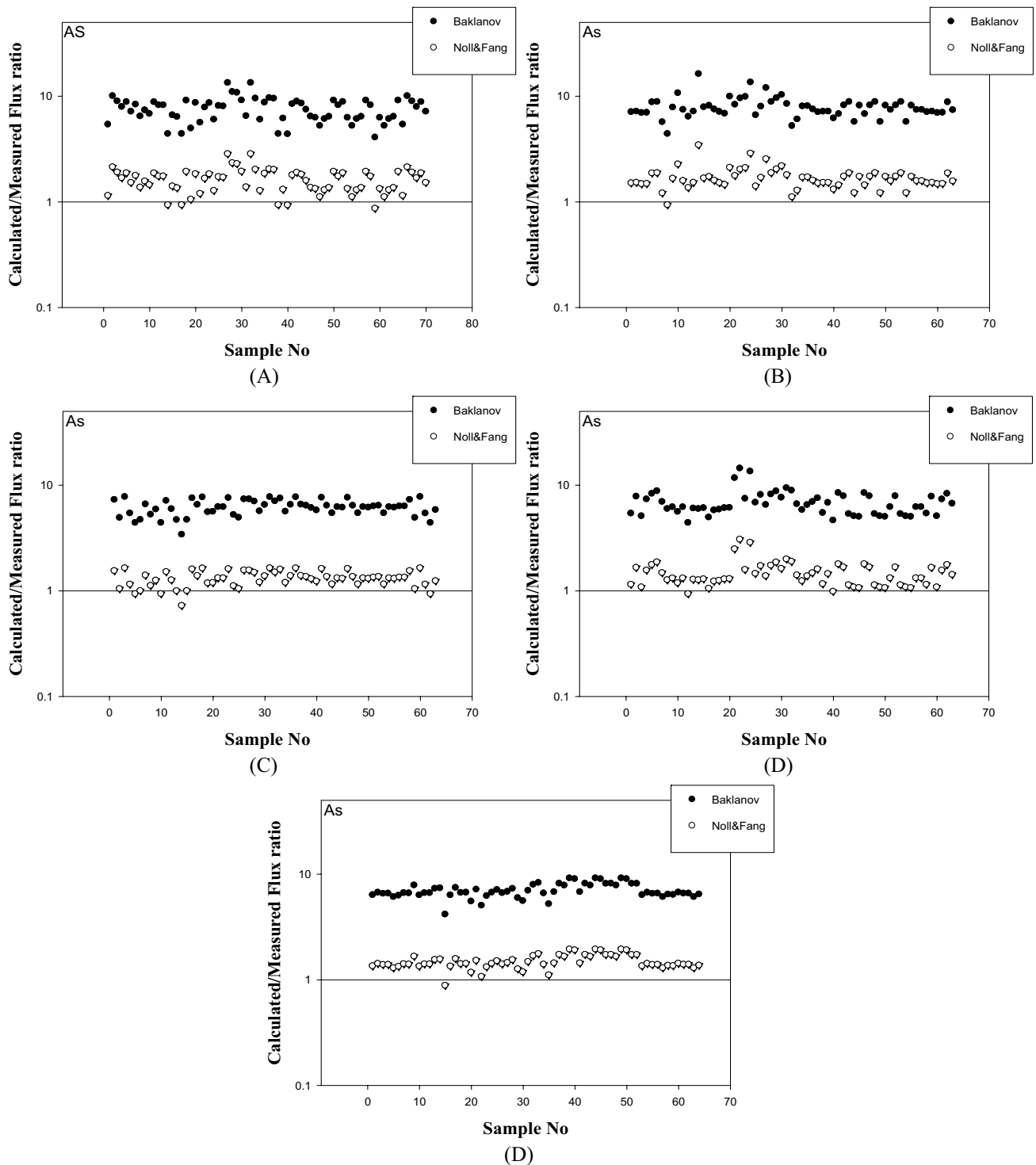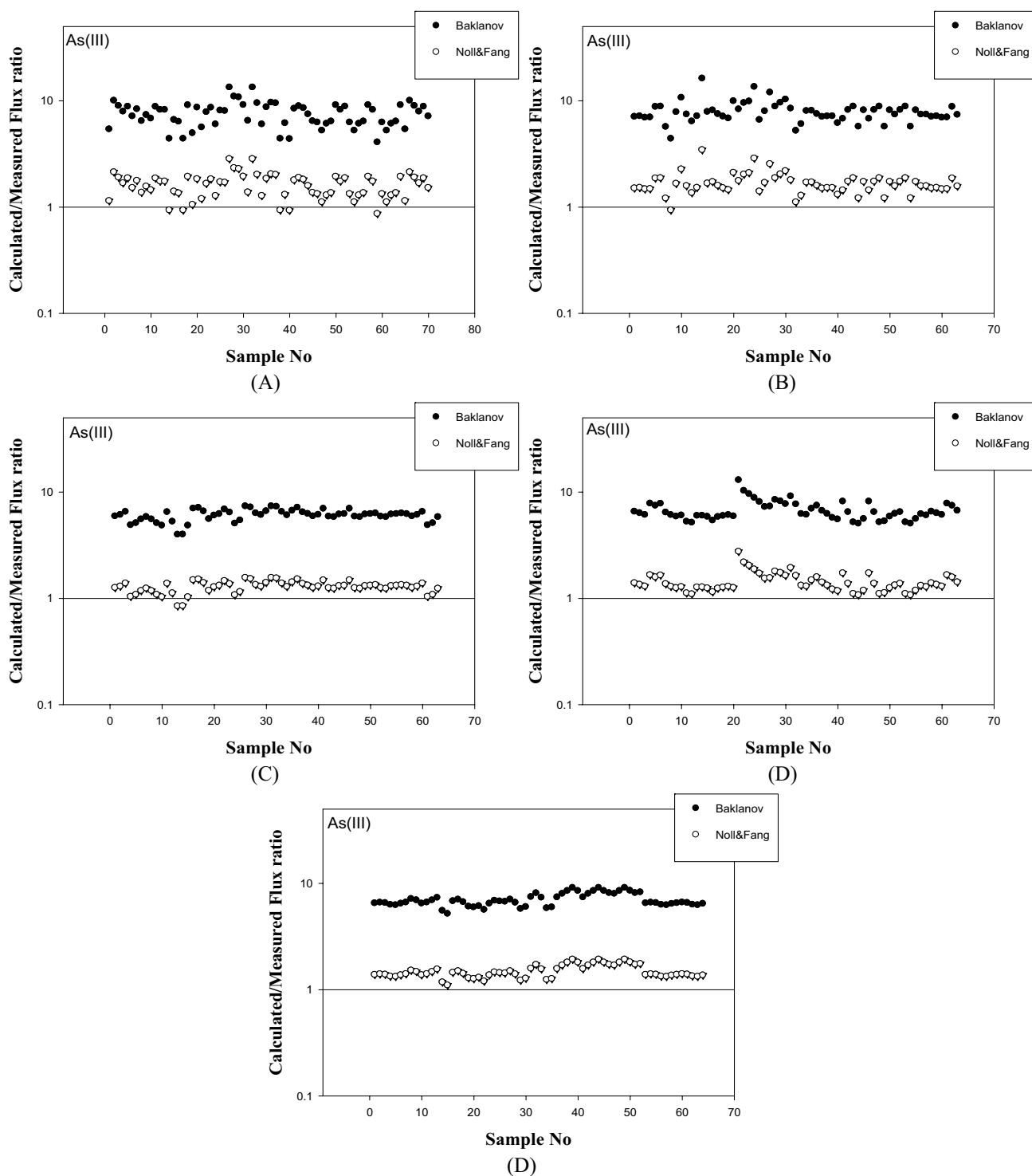
Fig. 2. Calculated/Measured flux ratios for arsenic (As) at five characteristic sampling sites (A) Bei-shi (suburban/coastal) (B) Chang-hua (downtown) (C) He-mei (residential) (D) Quan-xing (industrial) (E) Gao-mei (wetland) with two different deposition models.

mei, Quan-xing, and Gao-mei sites were 7.56, 7.91, 6.04, 6.72, and 6.98, respectively. Finally, the average calculated/ measured flux ratios for As(III) by the Noll and Fang model at the Bei-shi, Chang-hua, He-mei, Quan-xing, and Gao-mei sites were 1.60, 1.68, 1.28, 1.42 and 1.48, respectively.

Fig. 4 lists the calculated/measured flux ratios for As(V)

at five characteristic sampling sites (A) Bei-shi (suburban/ coastal) (B) Chang-hua (downtown) (C) He-mei (residential) (D) Quan-xing (industrial) (E) Gao-mei (wetland) using the two deposition models. Average calculated/measured flux ratios for As(V) by the Baklanov model at the Bei-shi suburban/coastal, Chang-hua downtown, He-mei residential,

Fig. 3. Calculated/Measured flux ratios for As(III) at five characteristic sampling sites (A) Bei-shi (suburban/coastal) (B) Chang-hua (downtown) (C) He-mei (residential) (D) Quan-xing (industrial) (E) Gao-mei (wetland) with two different deposition models.
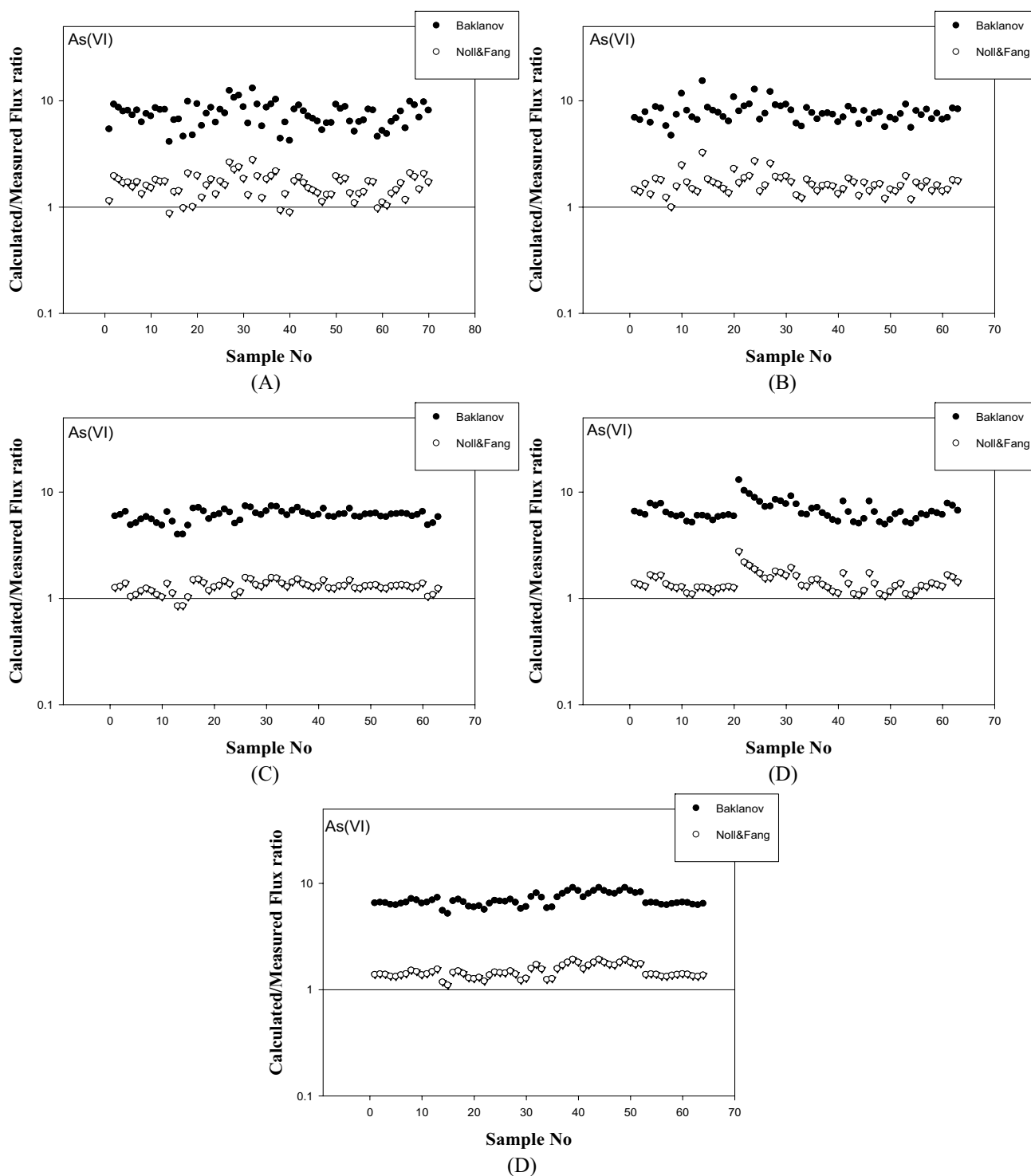
Quan-xing industrial, and Gao-mei wetland sites were 7.51, 7.83, 6.04, 6.68, and 6.98, respectively. Finally, average calculated/measured flux ratios for As(V) by the Noll and Fang model at the Bei-shi, Chang-hua, He-mei, Quan-xing, and Gao-mei sites were 1.59, 1.66, 1.28, 1.48, and 1.48, respectively.

CONCLUSIONS

The main conclusions are as follows.
1. Average concentrations of As, As(III), and As(V) in TSP during 2009–2010 in increasing order were the Quan-xing industrial site > Chang-hua downtown site

Fig. 4. Calculated/Measured flux ratios for As(V) at five characteristic sampling sites (A) Bei-shi (suburban/coastal) (B) Chang-hua (downtown) (C) He-mei (residential) (D) Quan-xing (industrial) (E) Gao-mei (wetland) with two different deposition models.

> Bei-shi suburban/coastal site > Gao-mei wetland site > He-mei residential site. Additionally, average dry deposition of As, As(III), and As(V) during the same period in increasing order was the Quan-xing industrial site > He-mei residential site > Bei-shi suburban/coastal site > Chang-hua downtown site >

Gao-mei wetland site.

2. Seasonal variations for As, As(III) and As(V) concentrations in TSP decreasing order were fall > winter > spring > summer at Bei-shi, Quan-xing and Gao-mei sampling sites. Seasonal variations in dry deposition of As, As(III) and As(V) in decreasing order

were fall > winter > spring > summer at Chang-hua, He-mei and Gao-mei sampling sites during 2009–2010. In addition, the average dry deposition velocity for ambient air total arsenic (As) for five characteristic sampling sites was 0.60 (cm/sec) at for Bei-shi sampling sites. As for Chang-hua, the average dry deposition velocity for ambient air total arsenic (As) was 0.57 (cm/sec). As for He-mei, the average dry deposition velocity for ambient air total arsenic (As) was 0.73 (cm/sec). As for Quan-xing, the average dry deposition velocity for ambient air total arsenic (As) was 0.67 (cm/sec). As for Gao-mei, the average dry deposition velocity for ambient air total arsenic (As) was 0.66 (cm/sec).

3. The Noll and Fang model was the only model that overestimated dry deposition of As, As(III) and As(V) from ambient air at the five sampling sites. However, it had better predictive performance than the other depositions models that were considered herein.

4. The calculated/measured flux ratio was fallen into an appropriate ratio indicating that the Noll and Fang model can be applied to predict dry deposition of As, As(III), and As(V) at these sampling sites.

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