



Mercury Emissions from a Coal-Fired Power Plant and Their Impact on the Nearby Environment

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ABSTRACT

This study investigated Hg emissions from a coal-fired power plant (CFPP) and their impact on the nearby environment. Atmospheric Hg concentrations were measured at sampling sites near a CFPP located in central Taiwan from November 2008 to March 2011. The mean gaseous and particulate Hg concentrations were 2.59–4.12 ng/m³ and 105–182 pg/m³, respectively, with gaseous Hg predominant at all sites (approximately 96% of the total atmospheric Hg). The seasonal variations of both gaseous and particle Hg concentrations in the atmosphere showed a similar pattern, with the highest concentrations in the cold season and the lowest in warm season. The mean emission factor of 13.1 mg/ton was found for the CFPP burning bituminous coal, with an electrostatic precipitator (ESP), flue gas desulfurization (FGD), and selective catalytic reduction (SCR) in series as air pollution control devices (APCDs). This figure was significantly lower than that measured at various power facilities, probably due to different fuel type, APCDs configuration, and flue gas condition. The modeling of the Industrial Source Complex Short Term (ISCST) revealed that the contribution of the CFPP to ambient atmospheric Hg was minimal (less than 1%).

Keywords: Mercury; Coal-fired power plant; Emission factor; Atmospheric emission.

INTRODUCTION

Mercury (Hg) compounds are highly toxic pollutants, and of great concern with regard to the global environment. Human health hazards from these pollutants are associated with their persistence in the environment and their potential bioaccumulation in the food chain (Fang *et al.*, 2010). The atmospheric emission of Hg mainly comes from anthropogenic and natural sources, and global emissions into the atmosphere via anthropogenic sources were approximately 1,930 tons in 2005, of which 45.6% has been directly attributed to thermal power plants followed by artisanal and small-scale gold production (18.2%) and metal production (10.4%) (AMAP/UNEP, 2008). Coal combustion

for power generation and residential heating is believed to make the most important contribution to atmospheric emissions (Pacyna *et al.*, 2006), and the Clean Air Mercury Rule was implemented by the US EPA in 2005 to control Hg emissions from coal-fired power plants (USEPA, 2005).

Mercury is present in coal in relatively low concentrations (approximately 0.1 ppmw), and is emitted into the environment at combustion temperatures above 150°C (Senior *et al.*, 2000; Zhang and Wong, 2007). In general, Hg compounds from coal combustion mainly consist of particle-bound Hg (Hg_p), gaseous elemental Hg (Hg⁰), and gaseous oxide Hg (Hg²⁺) (Galbreath and Zygarlicke, 2000). Hg⁰ is the most persistent form of Hg in the atmosphere (lifetime of 0.5 to 2 years) because of its low reactivity and solubility in water (Schroeder and Munthe, 1998), and thus it can be transported far from the immediate emission sources. Hg_p and Hg²⁺ have much shorter atmospheric lifetimes, measured in days or few weeks, due to their high reactivity and water solubility, which cause a faster deposition than Hg⁰ through both dry and wet processes

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(Schroeder and Munthe, 1998). Due to the special properties of Hg_p and Hg^{2+} , they can be efficiently controlled by conventional air pollution control devices (APCDs), such as electrostatic precipitators (ESP), flue gas desulfurization (FGD), and fabric filters (FF) (Zhang *et al.*, 2008). Wang (2010a) indicated that the Hg removal efficiency in combined ESP and FGD systems ranged from 24% to 72%.

In Taiwan, coal-fired power plants account for approximately 32.1% of total electricity generation, based on 2010 statistics (MOEA, 2010). Both coal and fuel oil combustion generate emissions of persistent organic compounds, including polychlorinated dibenzo-*p*-dioxins and-furans (PCDD/Fs), polybrominated diphenyl ethers (PBDEs), and polycyclic aromatic hydrocarbons (PAHs), as well as many other major pollutants (e.g., particulates, carbon, sulfur, nitrogen oxides, and Hg) (Lin *et al.*, 2007; Lin *et al.*, 2010; Vega *et al.*, 2010; Wang *et al.*, 2010b; Xue *et al.*, 2010; Bari *et al.*, 2011; Chen *et al.*, 2011; Tsai *et al.*, 2011). The total amount of Hg emissions from coal-fired power plants has thus emerged as an important issue due to the high volume of flue gas such facilities produce. However, little research has been carried out into the Hg emissions from coal-fired power plants and their impacts on the nearby atmospheric environment.

To address this gap in the literature, this study examines the fate and behavior of Hg from a coal-fired power plant (CFPP) located in central Taiwan. The impacts associated with Hg emissions from a coal-fired power plant on the nearby areas were assessed based on the Industrial Source Complex Short Term (ISCST) model, and the annual Hg contribution to these areas was then predicted.

MATERIALS AND METHODS

The Selected CFPP

The CFPP examined in this work consists of ten boilers with a capacity of 550 MW each. During the full load operation of the power plant, the amount of bituminous coal blend consumed by each boiler is about 213 tons/hour. The CFPP is installed with a selective catalytic reduction (SCR) (for NO_x control) followed by ESP and wet FGD (for SO_2 removal) as APCDs, and the treated flue gas is then emitted via a 250 m height stack.

Ambient Sampling Site

Five sampling sites with the maximum ground concentration of Hg from the CFPP emissions were found using the ISCST model. All sites are located between the Taiwan Strait and Taichung, and are briefly described as follows. Site A is on the rooftop of a service station located in an urban (industrial) area and is on the northeastern upwind side approximately 11 km from the CFPP. The site is also influenced by mobile sources from the highway (Formosa No. 3, about 1 km east of the site) and a municipal solid waste incinerator (MSWI) (Houli, 13 km northeast). Site B is on the southeastern downwind side about 2.3 km from the CFPP, and is on the rooftop of an elementary school located in a coastal suburban area. It is close to a steel plant, which is 2.7 km to the northwest. The remaining downwind

sites, C, D, and E, are on the rooftops of elementary schools located in rural areas and are on southern side about 7, 10, and 12 km from the CFPP, respectively. These sites are close to two MSWIs (Wenshan, 12 km northeast, and Wurih, 15 km southeast) and secondary aluminum smelters (around 10 km northeast). The locations of the selected CFPP, five sampling sites, and other possible Hg emission sources have been listed in our previous work (Wu *et al.*, 2010).

Sampling and Analysis

All samples were collected from the stack flue gas in accordance with US EPA Method 29. Particulate Hg (Hg_p) was collected in the probe and on a heated filter, gaseous Hg (Hg^0 and Hg^{2+}) was then collected in seven impingers with acidic solutions. Ambient air samples were collected simultaneously following US EPA Method IO-5. Gaseous Hg (Hg^0 and Hg^{2+}) was collected using two-stage gold amalgamation. A Teflon filter pack with a glass fiber filter was placed in front of the traps to remove particulate material from the air being sampled. Air was pulled through the gaseous sampling system using a mass-flow controlled vacuum pump at a nominal flow rate of 0.3 L/min. Particulate Hg (Hg_p) was collected on glass fiber filters using a vacuum pump at a nominal flow rate of 30 L/min. The recovered flue gas and ambient air samples were digested, and appropriate fractions were analyzed for Hg using dual gold amalgamation coupled with cold vapor atomic fluorescence spectrometry (CVAFS).

A total of 40 ambient Hg samples were collected over all four seasons from November 2008 to March 2011, and all meteorological information for the sampling sites during the investigation period is given in Table 1. The maximum and minimum temperatures at the sampling areas were 28.4°C (in summer) and 21.3°C (in winter). The prevailing wind directions varied with the season, although normally there were north and northwest winds. The wind speed ranged from 5.2 to 9.1 m/s, with the highest value found in August and September (i.e., in the fall).

ISCST Modeling

The ISCST is the current regulatory model approved by US EPA to estimate the ambient impacts of various emission sources out to distance of about 50 km (Lee *et al.*, 2009; Yu *et al.*, 2010; Tu *et al.*, 2011). Annual atmospheric mercury concentrations were predicted on a 400 m Cartesian grid of ground level positions in a 30 × 30 km area surrounding the investigated CFPP (a total of 10 stacks) using an ISCST model. The model was run under no buoyancy-induced dispersion conditions in a rural setting. The characteristics of the 10 stacks, such as height, diameter, temperature, flow rate, and exit velocity, were used to run the model (see Table 2). The meteorological data obtained from the Taichung Weather Service Office, together with stack parameters, were used as inputs for running the ISCST model on an annual basis.

RESULTS AND DISCUSSION

Ambient Hg Concentrations

Table 1. Meteorological information during the sampling period.

Season	Winter	Spring	Summer	Fall
Sampling period (dd/mm/yy)	25–26/11/08 21–22/12/10	24–25/01/09 01–01/03/11	07–08/07/09 06–07/07/10	25–26/08/09 14–15/09/10
Sampling number	10	10	10	10
Average Temperature (°C)	21.9	21.3	28.4	28.4
Relative humidity (%)	74	74	75	77
Average wind speed (m/s)	5.9	5.4	5.2	9.1
Direction of prevailing wind	North-northwest	East-southeast	North-northwest	North-northwest

Table 2. Stack characteristics and Hg emission rates.

Stack	Stack height (m)	Stack diameter (m)	Exhaust gas temperature (K)	Mean Hg concentration (ng/Nm ³)	Exit Velocity (m/s)	Hg emission rate (g/s)
1	250	6.7	379	1232	1232	5.81E-04
2	250	6.7	376	1038	1038	5.22E-04
3	250	6.7	374	1620	1620	7.67E-04
4	250	6.7	384	1604	1604	7.82E-04
5	250	6.7	371	1593	1593	8.25E-04
6	250	6.7	372	614	614	2.86E-04
7	250	6.7	374	1924	1924	9.58E-04
8	250	6.7	374	1829	1829	8.93E-04
9	250	6.7	370	2672	2672	1.42E-03
10	250	6.7	368	1366	1366	6.77E-04

The average gaseous and particulate Hg ranged from 2.59–4.12 ng/m³ and 105–182 pg/m³, respectively (Table 3). Gaseous Hg was the dominant species measured at all sampling sites, accounting for 96% of the total atmospheric Hg. The highest concentration of total (gaseous + particulate) atmospheric Hg was found in downwind site D (4.29 ng/m³), while the lowest level was found in upwind site A (2.70 ng/m³). As noted in the previous section, sampling site B was the one nearest to the CFPP, and hence its atmospheric Hg should mainly come from transient emissions of CFPP. If the transient emissions of CFPP were the predominant source for the study areas, a much higher level for site B than for the other downwind sites could be expected, although this was not found in this study. The results indicated that the highest Hg level of site D may be due to the various nearby emission sources, including MSWIs, electric arc furnaces, and non-ferrous metal smelting facilities, rather than the transient emissions of the CFPP (Liu *et al.*, 2002; Fu *et al.*, 2012), and all of these other sources are around 15 km north and northeast of the site. Since the prevailing winds during the sampling period mostly came from the north-northwest, agricultural waste open burning or some small point sources located in the north and northwest may also be significant Hg sources here. In addition, the Hg distribution may be affected by meteorological conditions, dry and wet depositions, and the coastal marine atmosphere (Sakata *et al.*, 2008; Fang *et al.*, 2010; Fu *et al.*, 2012).

The atmospheric gaseous Hg concentrations in this study (2.59–4.12 ng/m³) were 1.5–2.5 times lower than those observed at rural, suburban, and urban areas of Taiwan (6.10–15.8 ng/m³) (Kuo *et al.*, 2006), but similar findings

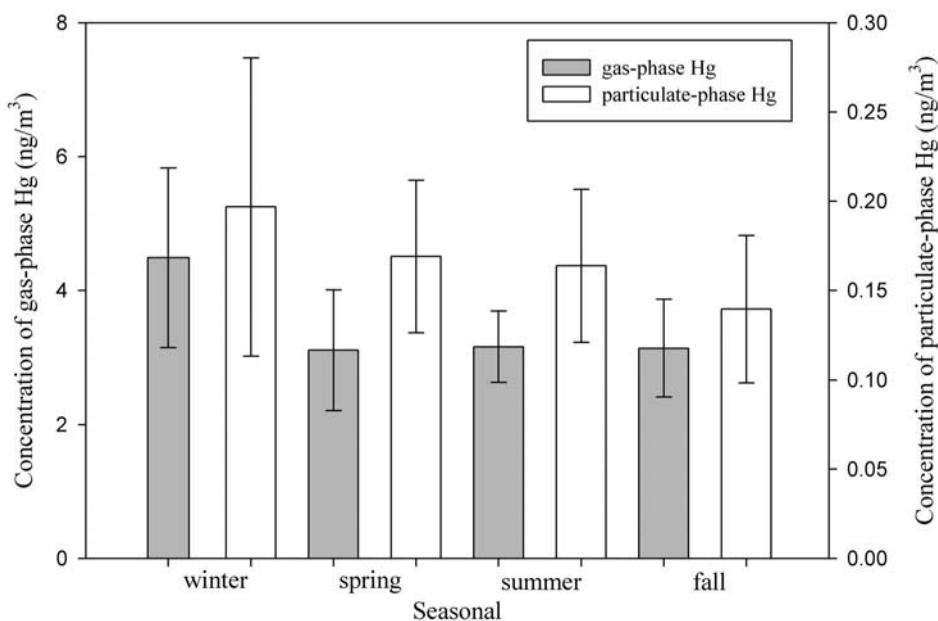
were observed at urban and suburban areas in Korea (3.22 ng/m³), Japan (2.88 ng/m³), and China (3.02–7.10 ng/m³) (Fu *et al.*, 2008; Kim *et al.*, 2009; Sakata and Marumoto, 2002). In addition, the gaseous Hg concentrations measured at low elevation sites in this study were significantly higher than the background levels measured at a high elevation station (Lulin, 2862 m a.s.l.) in Taiwan (0.240–5.75 ng/m³, mean = 1.73 ng/m³) due to the influence of local anthropogenic emission sources (Sheu *et al.*, 2010).

Seasonal Variations

Seasonal variations of average gaseous and particulate Hg are illustrated in Fig. 1. The data were collected from November 2008 to March 2011, which was broken down into winter (November–January), spring (February–April), summer (May–July), and fall (August–October). The mean values of gaseous and particulate Hg showed a similar seasonal pattern, with the highest concentrations in the cold season and lowest in the warm season (one-way ANOVA, $P < 0.05$). A significant correlation with the wind direction was also detected (one-way ANOVA, $P < 0.05$). Similar seasonal trends of atmospheric Hg were also observed at urban and suburban areas in China (Liu *et al.*, 2002; Fu *et al.*, 2012), and attributed to the residential activities (such as combustion of coal and biofuel for domestic heating). However, domestic heating is not required during winter in Taiwan, and thus can be neglected as a factor. The burning of agricultural residues has been shown to be a significant source of atmospheric Hg, and the emission factor of 0.037 g/ton-residue has been estimated in Oregon (Friedli *et al.*, 2003), which is higher than that of fuel oil (0.014 g/ton-oil) and biofuel combustion (0.020 g/ton-oil) (Streets *et al.*,

Table 3. Concentrations of gas and particulate phase Hg of the ambient sampling sites.

Sampling site	Gas phase Hg (ng/m ³)			Particulate phase Hg (pg/m ³)		
	Mean	Range	RSD (%)	Mean	Range	RSD (%)
A	2.59	1.46–3.93	36	105	64–139	30
B	3.44	2.24–6.01	22	140	97–227	22
C	3.71	2.58–5.28	21	182	100–256	33
D	4.12	3.56–6.20	9.8	171	130–303	17
E	3.49	2.68–5.37	21	169	109–316	25

**Fig. 1.** Seasonal variations of average concentrations of gaseous and particulate Hg.

2005). In Taiwan, around 1.3 Mt of rice and 0.65 Mt of rice straw are produced annually (COA, 2010). At harvest periods (June–July and November–December), most rice straw is usually dealt with by open field burning, resulting in a significant amount of Hg emissions. Since agricultural waste open burning is mostly during the cold season in central Taiwan, elevated atmospheric Hg in winter is likely related to these regional emissions. In addition, seasonal monsoon activities, dry deposition flux of Hg⁰, wet scavenging of Hg_p, and the boundary layer depth in winter are also important factors that might be related to the seasonal difference in atmospheric Hg concentrations (Fu *et al.*, 2008; Sheu *et al.*, 2010; Fu *et al.*, 2012).

Emissions of Hg from the Selected CFPP

The average Hg concentration measured at ten stack gases of the CFPP was 1.55 µg/Nm³ (ranging from 0.614 to 2.67 µg/Nm³) (Table 2). The emission rates of stack flue gases were calculated based on the Hg concentrations of flue gases and flow rates during the sampling period, and the calculated Hg emission rates ranged from 0.000286 to 0.00142 g/s. The average Hg concentrations in the flue gases were similar to those measured at a bituminous CFPP in Taiwan equipped with SCR, ESP, and FGD (0.29–1.25 µg/Nm³) (His *et al.*, 2010), and those from a bituminous coal consuming power plant in Korea with ESP and FGD

(1.03–2.41 µg/Nm³) (Lee *et al.*, 2006). However, the findings of this study were significantly lower than those reported in China with different APCDs. For example, the Hg concentrations measured at a CFPP equipped with ESP were found to 13–21 µg/Nm³ (Guo *et al.*, 2007) and 3.9–32 µg/Nm³ (Lee *et al.*, 2004), respectively. Lei *et al.* (2007) examined the Hg concentrations from six different CFPPs with various APCDs in China, and found that they ranged from 0.17 to 39.0 µg/Nm³. These results indicate that coal characteristics, type and efficiency of APCDs, and flue gas temperature are the major factors affecting the level of total mercury emissions to the atmosphere during the combustion of fuels (Lee *et al.*, 2004; Meij *et al.*, 2006; Shah *et al.*, 2008; Wang *et al.*, 2010a).

Hg Emission Factors

The emission factor is an important parameter for estimating the total emissions of Hg from a given source. In this study, the emission factor (13 mg/ton) for the CFPP was developed by multiplying the actual Hg concentration of the flue gas and the flue gas exhaust rate on a dry basis, and then dividing this by the coal burning rate. The emission factors of Hg from various thermal power plants are summarized in Table 4. The average emission factor for a CFPP burning bituminous coal is 13.1 mg/ton, which is close to the value (17.6 mg/ton) for bituminous coal-fired

Table 4. Emission factors of Hg from selected thermal power plants.

Country	Source	Type of APCDs	Emission factors (mg/ton)	Reference
Taiwan	Bituminous coal	SCR, ESP, FGD	13	This study
Korean	Bituminous coal	SCR, ESP, FGD	18	Kim <i>et al.</i> (2010)
	Anthracite	ESP	26	Kim <i>et al.</i> (2010)
China	Oil	ESP	3.3	Kim <i>et al.</i> (2010)
	Coal	ESP, FGD	238	Wang <i>et al.</i> (2000)
	Oil	-	65	Wang <i>et al.</i> (2000)
	Bituminous coal	ESP, FGD	43–75	Wang <i>et al.</i> (2010)
	Anthracite	ESP, FGD	32	Wang <i>et al.</i> (2010)
	Lignite	ESP, FGD	25	Wang <i>et al.</i> (2010)
	Bituminous coal	ESP, CFB-FGD, FF	131	Wang <i>et al.</i> (2010)
	Lignite	SCR, ESP, FGD	11	Wang <i>et al.</i> (2010)
United States	Bituminous coal	Uncontrolled	210	USEPA (1997)
	Anthracite	Uncontrolled	230	USEPA (1997)
	Subbituminous coal	Uncontrolled	100	USEPA (1997)
	Lignite	Uncontrolled	150	USEPA (1997)
Canada	Domestic bituminous coal	-	90	Innanen (1998)
	Imported bituminous coal	-	200	Innanen (1998)
	Lignite	-	130	Innanen (1998)
	Light fuel oil	-	50	Innanen (1998)
	Heavy fuel oil	-	410	Innanen (1998)
Europe	Coal	-	100–300	UN ECE (2000)
	Oil	-	1.0	UN ECE (2000)

Notes: (-), not specified; SCR, selective catalytic reduction; ESP, electrostatic precipitator; FGD, flue gas desulfurization; CFB-FGD, circulating fluidized bed FGD; FF, fabric filter.

boilers equipped with the same APCDs (SCR, ESP, and FGD) as reported in Korea (Kim *et al.*, 2010). However, the average emission factor in this study was 8–18 times lower than that of US plants without APCDs (USEPA, 1997), and was also significantly lower than that of coal-fired boilers with ESP and ESP + FGD as APCDs (Wang *et al.*, 2000; Kim *et al.*, 2010). The results revealed that the anthracite coal-burning boilers emitted a higher concentration of Hg than the bituminous coal-fired facilities, which is positively correlated with the Hg content of the coal. In addition, the emission factor is generally higher for coal-fired power plants than oil-fired power ones, due to higher content of Hg in coal rather than oil (Kim *et al.*, 2010). The differences in these data might be due to different types of fuel (coal or oil) and APCDs used.

Contributions of Hg to the Ambient Atmosphere

Based on the mean Hg concentration of the stack gases of the CFPP, the ambient atmospheric maximum hourly Hg concentrations at the sampling sites were obtained with the ISCST model. The maximum 10 1-hour average Hg concentrations were 3.32 to 3.58 ng/m³ (Table 5). These scenarios took place mostly in the afternoon (12:00–13:00 h) during the fall of 2009 (especially in August–September). The results indicated that the meteorological conditions in these periods may have contributed to high Hg concentrations found in the study areas.

The atmospheric Hg concentrations (ng/m³) contributed by the CFPP, as obtained with the ISCST model, are presented in Fig. 2. The maximum impact from the CFPP was

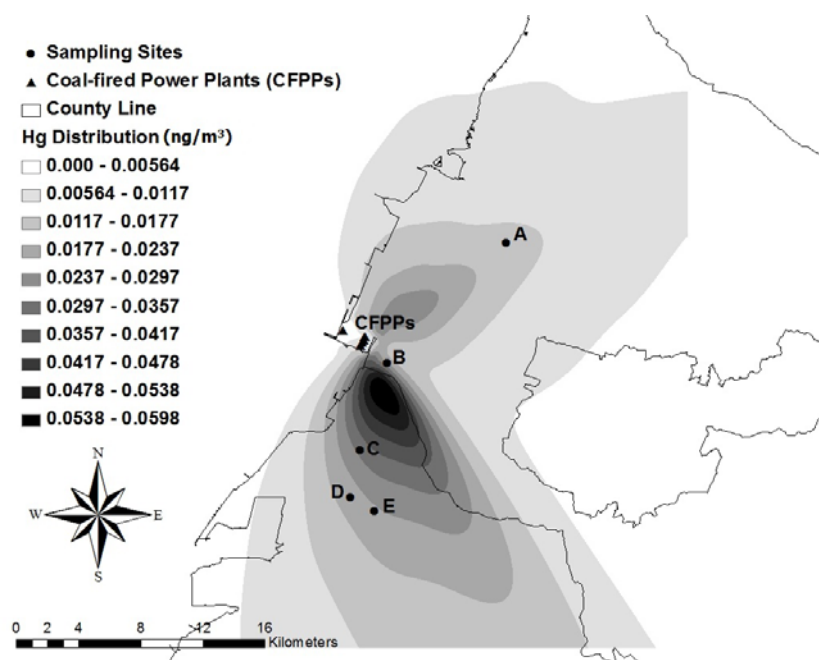
observed at approximately 5 km southeast of the CFPP, and this was highly correlated with the prevailing wind direction of the CFPP stack gases. Table 6 lists the measured gaseous Hg concentrations and the ISCST-modeled Hg concentrations. It should be noted that more than 99% of the Hg in the stack emissions was in gaseous form, and thus the proportion in particulate form was extremely low (Liu *et al.*, 2002). Therefore, we have considered only gaseous Hg when estimating the contribution of the CFPP to the ambient atmospheric Hg. Table 6 shows that the contribution of the CFPP to the ambient atmospheric Hg concentrations was quite low, with the Hg contribution fraction from the emission sources on the ambient air being less than 1% (mean = 0.648%, range = 0.505–0.851%). Consequently, the ambient atmospheric Hg concentrations in the study areas were not strongly influenced by the CFPP, probably because the larger stack height (250 m) and coastal marine atmosphere may lead to greater transportation of the emissions from the CFPP.

CONCLUSIONS

This study measured the average gaseous and particulate Hg at five sampling sites near a CFPP located in central Taiwan, and the results were 2.59–4.12 ng/m³ and 105–182 pg/m³, respectively. The maximum concentration of atmospheric Hg was found in downwind site D (4.29 ng/m³), which was highly associated with the prevailing wind direction of the CFPP stack gases. The results show that nearby emission sources, such as MSWIs, electric arc

Table 5. Maximum 1-hour average concentration of Hg calculated by using the ISCST model.

Rank	TWD97 coordinates		Concentration (ng/m ³)	Date (dd/mm/yy)	Time (hours)
	X	Y			
1	197891	2679727	3.58	26/11/09	1300
2	197891	2678127	3.56	26/08/09	1200
3	197891	2678127	3.52	21/03/09	1100
4	198291	2678927	3.52	26/05/09	1000
5	198291	2679327	3.52	26/08/09	1300
6	198291	2678927	3.38	16/09/09	1200
7	198291	2679327	3.38	03/09/09	1400
8	197491	2677727	3.36	17/09/09	1200
9	198291	2679727	3.35	14/08/09	1500
10	198291	2678527	3.32	25/11/09	1300

**Fig. 2.** Atmospheric Hg concentration (ng/m³) contributed by the CFPP, obtained by using the ISCST model.**Table 6.** Contribution of the CFPP to atmospheric Hg.

Sampling site	Mean gas-phase Hg concentration measured (ng/m ³)	ISCST3-modeled maximum hourly Hg concentration (ng/m ³)	Contribution by CFPP (%) (modeled concentration/measured concentration × 100%)
A	2.59	0.0135	0.523
B	3.44	0.0244	0.709
C	3.71	0.0316	0.851
D	4.12	0.0208	0.505
E	3.49	0.0228	0.653

furnaces, and non-ferrous metal smelting facilities, had a significant great impact on the study areas. A mean emission factor of 13.1 mg/ton was observed in this study, and that Hg emission was closely related to fuel type, APCDs configuration, and flue gas condition. The contributions of the CFPP to the ambient atmospheric Hg concentrations estimated by using ISCST model were quite low, with the Hg contribution fraction from the emission sources on the ambient air being less than 1%.

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