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Distribution, sources and health risk assessment of mercury in kindergarten dust

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HIGHLIGHTS

- The concentrations, sources and health risk of Hg in kindergarten dust were investigated.
- The emissions from coal-power plants were the main source of THg in the dust.
- Landfills play an important role in MeHg accumulation in the dust.
- Hg in the dust from educational area posed a high non-cancer risk on the children health.

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ABSTRACT

Mercury (Hg) contamination in urban area is a hot issue in environmental research. In this study, the distribution, sources and health risk of Hg in dust from 69 kindergartens in Wuhan, China, were investigated. In comparison with most other cities, the concentrations of total mercury (THg) and methylmercury (MeHg) were significantly elevated, ranging from 0.15 to 10.59 mg kg^{-1} and from 0.64 to 3.88 μg kg^{-1}, respectively. Among the five different urban areas, the educational area had the highest concentrations of THg and MeHg. The GIS mapping was used to identify the hot-spot areas and assess the potential pollution sources of Hg. The emissions of coal-power plants and coking plants were the main sources of THg in the dust, whereas the contributions of municipal solid waste (MSW) landfills and iron and steel smelting related industries were not significant. However, the emission of MSW landfills was considered to be an important source of MeHg in the studied area. The result of health risk assessment indicated that there was a high adverse health effect of the kindergarten dust in terms of Hg contamination on the children living in the educational area (Hazard index (HI) = 6.89).

1. Introduction

Mercury (Hg) is a ubiquitous environmental toxicant which can cause a wide range of adverse health effects in humans, especially children (Schuster et al., 2002; Gustin, 2003; Mather and Pyle, 2004). Hg vapor, for instance, can enter bloodstreams, be distributed to many tissues, such as liver, hear muscle, and brain, and be retained in human body (Li et al., 2008; Farzin et al., 2008). The adverse effects caused by Hg vapor are usually on central nervous system and kidneys, exhibiting emotional instability, gingivitis, tremors, and kidney failure (WHO, 1996). There is an increasing concern on the potential neurotoxic effect of chronic low-dose mercury exposure on children, an increasing global problem (Ip et al., 2004; Grandjean et al., 2004).

In atmosphere, mercury exists mainly in gaseous phase, unlike other heavy metal pollutants that tend to exist as particulate phase (Fu et al., 2012; Valente et al., 2007; Chand et al., 2008). Atmospheric Hg can be divided into gaseous elemental mercury (GEM)-Hg^0, reactive gaseous mercury (RGM)-Hg^{2+}, and particulate mercury (PM) (Bergan et al., 1999; Schroeder and Munthe, 1998; Streets et al., 2005; Pacyna et al., 2006a,b; 2010; Wu et al., 2006; Fu et al., 2012). RGM and PM can be rapidly removed from the atmosphere by wet and dry depositions, whereas GEM is only affected by dry

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deposition (Lee et al., 2001; Wängberg et al., 2005). GEM is quite stable with a lifetime of 0.5–2 years and is thus capable of distribution on a regional scale. On the contrary, RGM and PM are more readily deposited close to the emission sources (Grangeon et al., 2012). In China, GEM accounts for 56% of the total anthropogenic Hg emissions, whereas RGM and PM account for 32% and 12%, respectively, of the total Hg emissions (Streets et al., 2005).

The total mercury emissions to atmosphere in China have been estimated to be 500–700 tons per year (Fu et al., 2012). Approximately 45% derived from non-ferrous metals smelting, 38% from coal combustion, and 17% from miscellaneous activities (including chlor-alkali production, battery and fluorescent lamp production and cement production) (Streets et al., 2005). In urban environment, chlor-alkali plant and coal-power plant are most common sources of Hg pollutant (Biester et al., 2002; Pacyna et al., 2006a,b; Streets et al., 2005; Tian et al., 2010).

Atmospheric deposition is an important pathway for Hg that scavenged from atmosphere (Zhang and Wong, 2007), and soil and dust are the main resting places of the deposited Hg. In comparison with urban soils, dust is more easily to be contacted by urban inhabitants, especially children (Hu et al., 2011). Hg contaminants in dust can enter human bodies by inhalation, ingestion and dermal contact, causing a number of health problems (Lu et al., 2009). Kindergarten is the main venue of the urban children. However, very limited information is available on Hg in kindergarten dust of previous studies.

In the present study, the distribution, sources and health risk of Hg of kindergarten dust from Wuhan, central China, were investigated. The results will be useful for regulators and engineers in environmental planning and development of effective environmental management.

2. Materials and methods

2.1. Study area

Wuhan (29°58′–31°22′N, 113°41′–115°05′E), the capital of Hubei Province, is located in the middle reaches of the Yangtze River with an urban population of about 10.02 million in 2011. It is one of the biggest hubs for land, water and air transportation in China. Its climate represents a typical subtropical humid monsoon with an average annual temperature of 17.7 °C and average annual rainfall of 1300 mm. The prevailing wind is from northeast to southwest in winter and from south to north in summer. The number of kindergartens in the main city is 638 with children totally of 125, 351 (WMBS, 2009). The major industries in Wuhan include ferrous smelters, chemical plants, coal-power plants, papermaking, machine manufacturing, and auto industries.

2.2. Sample collection

Sixty-nine kindergarten dust samples were collected from five urban areas of commercial area (CA), educational area (EA), industrial area (IA), residential area (RA) and control district (CD). Sampling was undertaken in dry periods when no rain had occurred during the previous week. The coordinates of the sample location were recorded with a GPS, and the sampling locations were shown in Fig. 1. About 50 g dust sample was collected using polyethylene brush on impervious surface (children activities equipment, pavement and windowsill) in outdoor from five to eight points of each kindergarten. All dust samples were stored in sealed polyethylene bags, labeled and then transported to the laboratory.

2.3. Sample preparation and analysis

The dust samples were air dried at room temperature. The impurities such as stones and tree leaves were removed and then the samples were sieved through a 1.0 mm mesh nylon sieve by gently crushing aggregates without grinding sand-sized mineral fragments. In order to facilitate the digestion, the dust samples were ground with agate mortar to pass through a 0.075 mm nylon sieve for Hg species analysis (Yang et al., 2011).

300–500 mg sample was oxidized with 5 ml concentrated HNO_3 + HCl (1:3 v/v) in a Teflon vial in a Microwave oven (MDS2000, from CEM, USA) for 50 min. The digested solution was

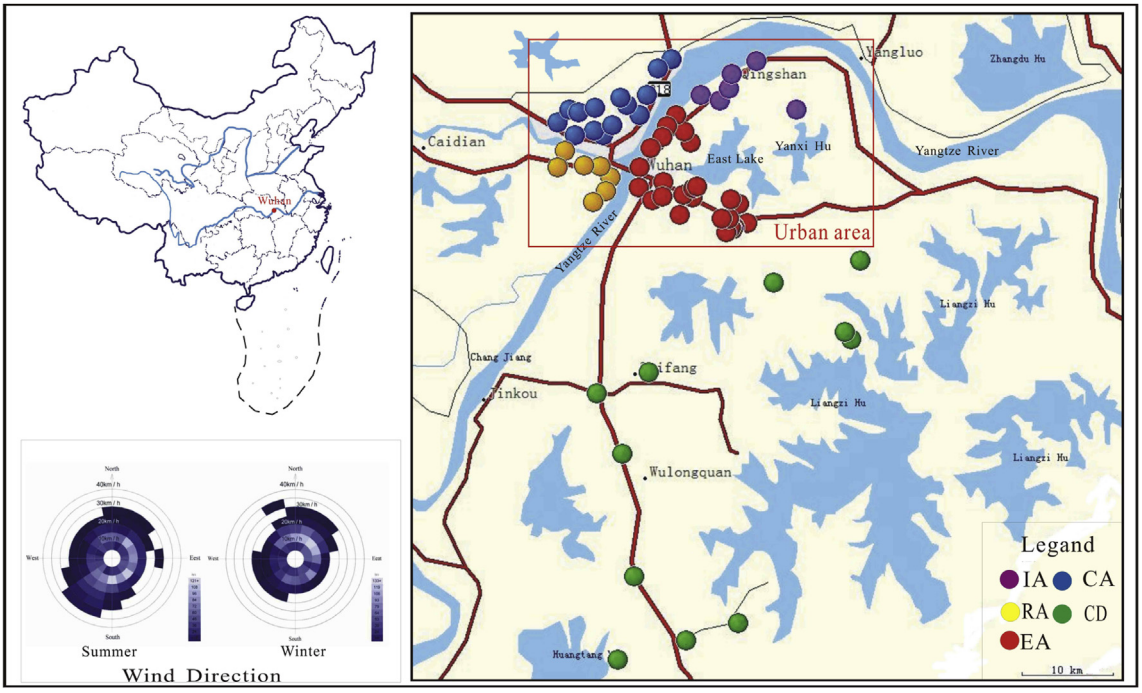

Fig. 1. Sampling sites and wind rose in the study area.

transferred to a 100 ml volumetric flask, and made up to 100 ml by adding Mill-Q water (Feng et al., 2006). Total mercury concentration was determined using BrCl oxidation and $SnCl_2$ reduction coupled with cold-vapor atomic absorption spectrometry (CVAAS) (Feng and Hong, 1999). For methylmercury analysis, prepared dust samples were digested using the $CuSO_4$-methanol/solvent extraction technique (Liang et al., 1994, 1996), and were measured by aqueous ethylation, purge, trap, and gas chromatography–cold vapor atomic fluorescence spectrometer (GC–CVAFS) according to USEPA Method 1630 (USEPA, 2001a).

Quality assurance and quality control were determined by method blanks, duplicates, and certified reference materials (SRM 2710 and GBW 07405 for THg; IAEA405 for MeHg). The relative percentage difference of sample duplicates was <10% and spike recoveries for Hg species analysis were between 86 and 109%.

2.4. Risk assessment model

Children expose to dust Hg via four main paths: (a) direct ingestion of substrate particles (D_{ing}); (b) inhalation of resuspended particles through mouth and nose (D_{inh}); (c) dermal absorption of mercury in particles adhered to exposed skin (D_{dermal}), and (d) exposure through inhalation of mercury vapor (D_{vapor}). The dose received through each of the four pathways can be calculated using Eqs. (1)–(4), which were adopted from U.S. Environmental Protection Agency (USEPA, 1989, 1996) and Zheng et al. (2010a, 2010b).

$$D_{ing} = C \times \frac{IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (1)$$

$$D_{inh} = C \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT} \quad (2)$$

$$D_{dermal} = C \times \frac{SL \times SA \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (3)$$

$$D_{vapour} = C \times \frac{InhR \times EF \times ED}{VF \times BW \times AT} \quad (4)$$

Table 1
Parameter values in average daily dose calculation models of mercury for children.

Parameter	Physical meaning	Units	Value	Date source
D	Daily intake dose	mg kg^{-1} day^{-1}		
C	Exposure-point concentration	mg kg^{-1}	95% UCL	This study
IngR	Ingestion rate	mg day^{-1}	200	USEPA (2001b)
InhR	Inhalation rate	m^3 day^{-1}	7.6	Van den Berg (1995)
EF	Exposure frequency	day a^{-1}	250	
ED	Exposure duration	a	6	USEPA (2001b) De Miguel et al. (2007)
BW	Average body weight	kg	15	USEPA (1989)
AT	Averaging exposure time	day	ED × 365	
PEF	Perticle emission factor	m^3 kg^{-1}	1.36 × 10^9	USEPA (2001b)
SL	Skin adherence factor	mg cm^{-2} day^{-1}	0.2	USEPA (2001b)
SA	Exposed skin area	cm^2	1150	Wang et al. (2008)
ABS	Dermal absorption factor	Unitless	0.001	Ferreira-Baptista and De Miguel (2005)
VF	Volatilization factor	m^3 kg^{-1}	32675.6	USEPA (2001b)

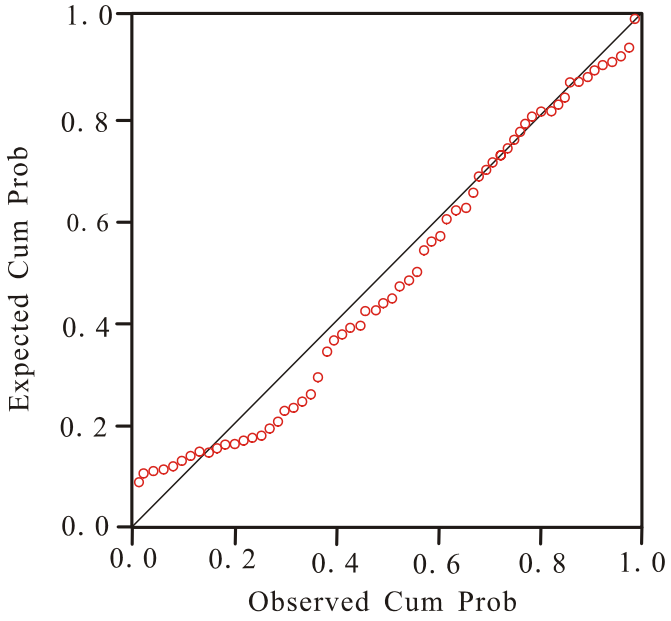

Fig. 2. P–P plot of THg in kindergarten dust of Wuhan.

It is assumed that children stay at home with parents in weekends and holidays and stay at kindergartens in the rest of the year. So we used 250 days per year as the average exposure frequency (EF). The other exposure parameters are listed in Table 1. *C* (exposure-point concentration, mg kg^{-1}) in Eqs. (1)–(4), combined with the values for the exposure factors shown above, is considered to yield an estimate of the "reasonable maximum exposure" (USEPA, 1989), which is the upper limit of the 95% confidence interval for mean (95% UCL) (USEPA, 1996). Since the concentrations of mercury in this study follow an approximate log-normal distribution (Fig. 2), the 95% UCL was calculated using the statistical software (SPSS 15.0 for Windows).

Hazard index (HI) (also stated as hazard quotient) was calculated according to Eq. (5) (Guney et al., 2010):

$$HI_i = \frac{D_i}{RfD_i} \quad (5)$$

where *RfD* is reference dose (mg kg^{-1} day^{-1}) and *i* is route of exposure. Hazard index (HI) for non-carcinogenic effects was applied to each exposure pathway in the analysis. The approach assumes that simultaneous subthreshold exposures to mercury could result in adverse health effects and the magnitude of the

Table 2
THg and MeHg concentrations in kindergarten dust of Wuhan.

	C_{THg} (mg kg^{-1})	C_{MeHg} (μg kg^{-1})
Number	69	69
Max	10.59	3.88
Min	0.15	0.64
Mean	1.66	1.57
Median	0.58	1.43
S.D.	2.32	0.71
C.V.	1.39	0.45
Skewness	2.03	1.52
Kurtosis	3.67	2.49
Guidelines[a]	1.0	NA
Wuhan BK[b]	0.032	NA

NA: not available.
[a] Class II, Environmental Quality Standard for soils (GB 15618-1995).
[b] Background values of the soil in Wuhan (Yang and Dong, 2004).

Table 3
THg and MeHg concentrations in dust of other cities reported in the literature.

Land type	Location	THg (mg kg^{-1})		MeHg (μg kg^{-1})		Reference
		Mean	Range	Mean	Range	
Kindergarten dust	Wuhan, China	1.66	0.15–10.59	1.57	0.64–3.88	This study
Street dust	Beijing, China	0.34	0.045–1.378			Zhang et al. (2006)
Street dust	Xiamen, China	0.28	0.034–1.4	0.55	0.092–2.3	Liang et al. (2009)
Street dust	Nanjing, China	0.12	0.05–0.34			Hu et al. (2011)
Zinc smelting district	Huludao, China	1.222	0.119–5.212			Zheng et al. (2010)a,b
Roadway dust	Baoji, China	1.11	0.48–2.32			Lu et al. (2009)
Street dust	Luanda, Angola	0.13	0.03–0.57			Lu et al. (2009)
Street dust	Avile's, Spain	2.56	1.20–10.8			Lu et al. (2009)

adverse effects will be proportional to the sum of the ratios of the subthreshold exposures to acceptable exposures (USEPA, 1989). If the value of HI is less than one, it is believed that there is no significant risk of non-carcinogenic effects. If HI exceeds one, then there is a chance that non-carcinogenic effects occur, with a probability which tends to increase as the value of HI increases (USEPA, 2001b).

3. Results and discussion

3.1. Mercury concentrations

Descriptive statistics of total mercury (THg) and methylmercury (MeHg) concentrations in the dust from kindergartens in Wuhan, as well as local background values of soils (Yang and Dong, 2004) and the guidelines value for soils in China (NEPA, 1995) are summarized in Table 2. The skewness of THg and MeHg were 2.03 and 1.52, respectively, revealing a strongly positively skewed distribution, which indicated the existences of some highly contaminated spots (Chen et al., 2010). In addition, the high standard deviations and coefficient of variation (CV) indicated the high inhomogeneity of both THg and MeHg in dust from the present study (Table 2). The concentrations of THg in the dust had a range of 0.15–10.59 mg kg^{-1}, with a mean of 1.66 mg kg^{-1} and a median of 0.58 mg kg^{-1}, which were about 52 and 18 times, respectively, higher than the background value of the soils (0.032 mg kg^{-1}). Similarly, the concentrations of THg were much higher than the guidelines value for soils. This result clearly indicated a great enhancement of Hg in the dust. The concentrations of MeHg ranged from 0.64 to 3.88 μg kg^{-1} with a mean value of 1.57 μg kg^{-1} and a median of 1.43 μg kg^{-1}. The ratios of MeHg/THg were in the range of 0.004–1.42%, with a mean of 0.20%, which was similar to a previous study (0.28%) for road dust of Xiamen, China (Liang et al., 2009).

The concentrations of THg and MeHg in dust reported in literatures are listed in Table 3. In comparison with the cities of Beijing, Xiamen, Nangjing, Huludao, Baoji, and Luanda, the concentrations of THg in the dust of kindergartens in Wuhan were much higher. At the same time, the mean concentration of MeHg in current study was higher than that of Xiamen. The elevated Hg in the dust might reveal simultaneously high values of Hg in the atmosphere of the studied city. Indeed, the total gaseous mercury in Wuhan had a mean concentrations of 14.8 ng m^{-3} (Xiang and Liu, 2008), which was much higher than those of Guiyang (9.7 ng m^{-3}), Shanghai (2.7 ng m^{-3}) and Chongqing (6.7 ng m^{-3}) (Fu et al., 2012).

The distribution of THg and MeHg in different urban areas of Wuhan are showed in Table 4. The concentrations of THg and MeHg with different types of land use were in the following decreasing order of EA > RA > IA > CA > CD and EA > CA > CD > IA > RA, respectively. The mean concentrations of THg and MeHg in Educational area were significantly higher than that from the other areas ($p < 0.05$, Two Independent Samples Test), while there were no significant differences among the areas of RA, IA, CA, and CD ($p > 0.05$). The higher values of Hg in EA might be attributed to the emissions of coal-power plant and coking plant located adjacent to this area (see discussion below). Despite the control district is located in the suburb and is far away from the main pollution sources, the concentrations of Hg in the dust from this area were not significantly lower than those from the other urban areas. The relatively intensive use of household coal might be an important source of Hg in the control district.

3.2. Sources identification

The geochemical map of mercury overlapping with the distribution of the major known anthropogenic pollution sources might reveal the possible sources of Hg in the dust. As shown in Fig. 3, two obvious "hotspots" were found distributing in the center (I) and south-east (II) parts of the city. These hotspots matched well with the locations of the coal-power plants and coking plant, and it thus could be concluded that the high concentrations of THg in dust from these areas were mainly derived from the emissions of these plants. It is evident that coal combustion is one of the most important Hg emission sources (Pacyna et al., 2006a,b; Streets et al., 2005; Tian et al., 2010), which accounts for about 20% of the total anthropogenic Hg emission in the studied city (Statistic Bureau of Wuhan, 1990–2011).

In addition, there was an area with slightly elevated THg concentrations distributing in the west part of the city (Fig. 3). Despite a municipal solid waste (MSW) landfill located close to this area (Fig. 3), it might not be the main cause of the elevated Hg in the dust. Li et al. (2010) had investigated the Hg contamination situation of this landfill and found that the concentrations of THg in the MSW and the cover soils were only 0.606 and 0.058 mg kg^{-1}, respectively. Furthermore, the dust samples collected near another larger MSW landfill (in the north part of the city) (Fig. 3) showed no similarly elevated THg concentrations. The elevated THg in the west part of the city was in agreement with previous studies which had also found high concentrations of Hg in soils (Gong

Table 4
Descriptive statistics of THg and MeHg concentrations in 5 functional areas in Wuhan.

Land function	Sample number	C_{THg} (mg kg^{-1})			C_{MeHg} (μg kg^{-1})		
		Range	Mean ± SD	CV	Range	Mean ± SD	CV
Commercial area	15	0.17–1.48	0.35 ± 0.33	0.93	1.15–2.63	1.56 ± 0.71	0.46
Educational area	27	0.55–10.59	3.44 ± 2.70	0.78	0.98–3.88	1.67 ± 0.66	0.39
Residential area	8	0.15–1.61	0.49 ± 0.48	0.97	0.97–1.43	1.16 ± 0.24	0.21
Industrial area	6	0.21–0.76	0.45 ± 0.25	0.55	0.64–2.26	1.33 ± 0.74	0.53
Control district	13	0.15–0.47	0.27 ± 0.10	0.37	0.64–3.54	1.43 ± 1.10	0.77

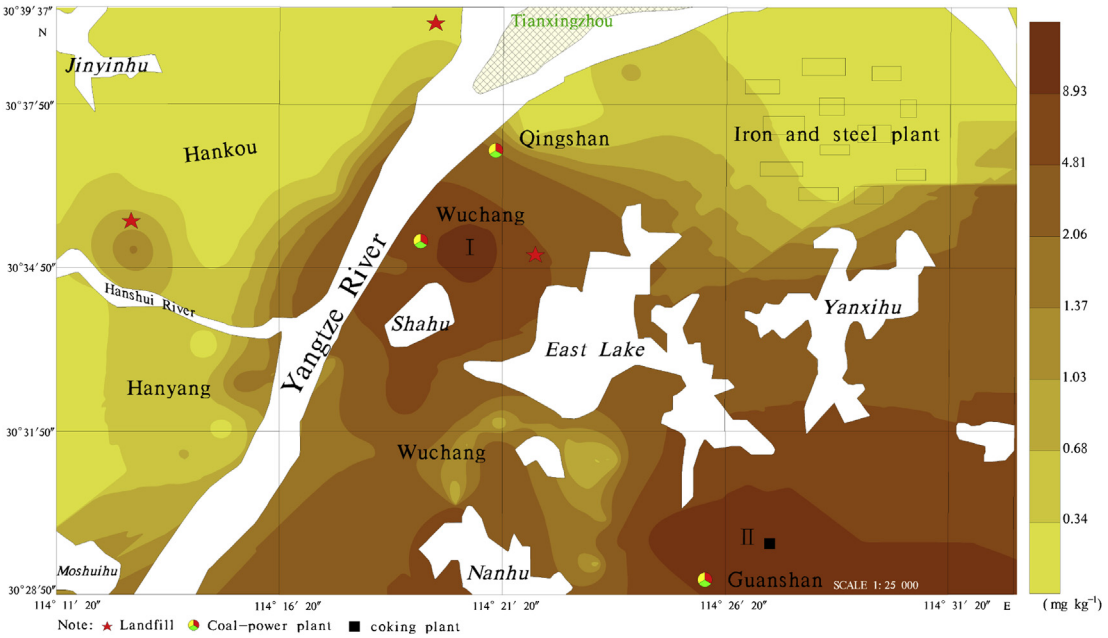

Fig. 3. GIS map of THg in kindergarten dust of Wuhan.

et al., 2010) and atmosphere (Xiang and Liu, 2008) from the same area, and this might be caused by a site specific pollution sources, such as the small chemical plants in this area (Xiang and Liu, 2008).

The spatial distribution of MeHg in the dust was similar to the THg with high values existing around the coal-power plants and coking plant (Fig. 4), which suggested that the levels of MeHg in the dust from these areas depended on the THg concentrations. However, there was also an area with obviously elevated concentrations of MeHg around a MSW landfill located in the north part of studied city (Fig. 4). This demonstrated that landfill could be an important source of methylmercury in the dust. This result was in agreement with previous studies which reported that landfill sites were effective bioreactors that converted inorganic mercury into methylmercury (Li et al., 2010; Feng et al., 2004).

It should be noted that the dust samples collected from the industrial area (iron and steel plants and related industries) showed no high concentrations of Hg (Figs. 3 and 4), which was quite different from other metal pollutants, such as Cu, Pb, and Zn (Gong et al., 2010). This reflected that the industrial activities (iron and steel smelting and processing) were not important sources of Hg in the dust.

The total amounts of Hg emissions from the main pollution sources in the urban area of Wuhan from 1990 to 2011 were calculated and showed in Fig. 5. It's obvious that coal combustion and coke production are the main emission sources of Hg in

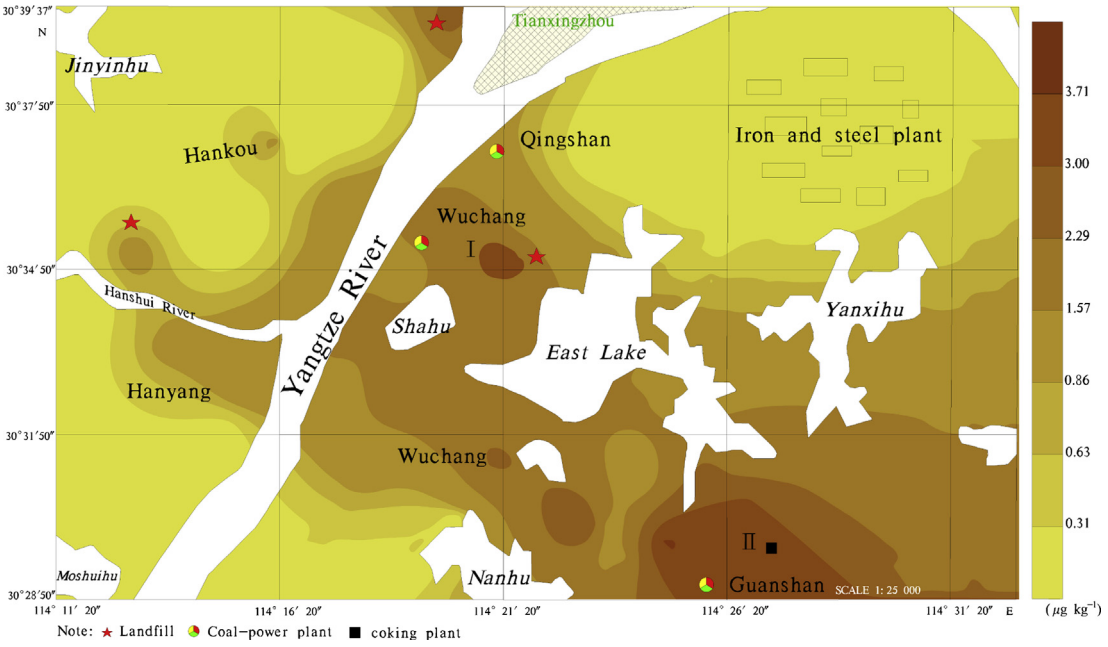

Fig. 4. GIS map of MeHg in kindergarten dust of Wuhan.

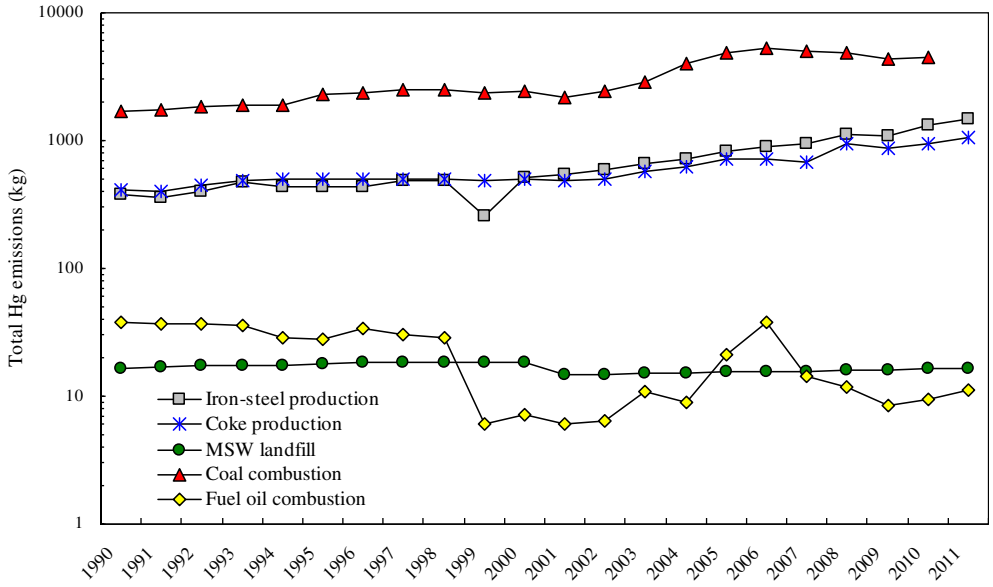

Fig. 5. Trend of total Hg emissions from different sources in Wuhan, China, 1990–2011. The total amounts of Hg emissions from the main pollution sources were calculated based on the *emission* factors (Wang et al., 2006; Streets et al., 2005; Wang et al., 1999; Jiang et al., 2005; UNEP, 2005) and the production or consumption of products or energy (Statistic Bureau of Wuhan, 1990–2011) of each source annually.

Wuhan, whereas the contributions of MSW landfill and fuel oil combustion are insignificant. This is quite in agreement with the result of the spatial distribution of Hg in the dust. However, the fact that iron and steel smelting can also release substantial amount of Hg (Fig. 4) is contradictory to the low levels of Hg in the dust collected around the iron and steel smelting industrial area (Fig. 3). Previous studies also found relatively low levels of Hg in the atmosphere (Xiang and Liu, 2008) and soils (Gong et al., 2010) from the same industrial area. This contradiction might be attributed to the use of advance systems for desulphurization and dust removal in these industries (Du et al., 2002).

3.3. Risk assessment

The results of risk assessment of dust Hg to children in Wuhan are listed in Table 5. Reference doses for mercury used in the analysis were taken from the US Department of Energy's RAIS compilation (2004), which are 3.0E−4 for direct ingestion, 2.10E−5 for dermal absorption and 8.57E−5 for inhalation through not only mouth and nose, but also vapor.

For non-cancer effect, in the case of children, inhalation of mercury vapor is the main route of exposure in all of the exposure pathways, which was consistent with the study of Fang et al. (2011) and Meza-Figueroa et al. (2007). This is due to the significant vapor pressure of Hg at ambient temperature (Meza-Figueroa et al., 2007).

The HIs for different functional areas of the studied city were in the order of CD < CA < IA < EA < EA (Table 5). Regarding non-cancer effects, Hazard indexes of inhalation and direct ingestion for educational area were larger than safe level one (6.86), which indicated a high health risk of Hg on the children living in this area.

Table 5
Hazard index (HI) for Hg and exposure pathway in different function areas.

Land function	HI_{ing}	HI_{inh}	HI_{dermal}	HI_{vapor}	$\sum HI_i$
Commercial area	1.63E − 02	1.60E − 03	2.69E − 04	6.65E − 02	8.47E − 02
Educational area	1.32E + 00	1.29E − 01	2.17E − 02	5.38E + 00	6.86E + 00
Residential area	2.62E − 02	2.56E − 03	4.30E − 04	1.07E − 01	1.36E − 01
Industrial area	2.15E − 02	2.11E − 03	3.54E − 04	8.76E − 02	1.12E − 01
Control district	1.01E − 02	9.91E − 04	1.66E − 04	4.12E − 02	5.25E − 02

It should be noted that this assessment only considered one urban medium (dust). In fact, Hg pollutants can be introduced into human bodies via various compartments, such as food, water, and atmosphere. Therefore, the exact HIs for the inhabitants should be higher. Furthermore, children living close to certain mercury pollution sources, such as coal-power plant and coking plant, would experience extremely adverse health effects. Therefore, the ecological and health effects of the enriched Hg in the studied city need further detailed investigations.

4. Conclusions

The concentrations of THg and MeHg in the dust collected from kindergartens in Wuhan, China ranged from 0.15 to 10.59 mg kg^{-1} and from 0.64 to 3.88 μg kg^{-1}, respectively. The distributions of THg and MeHg among the five different urban areas were in the following decreasing order of EA > RA > IA > CA > CD and EA > CA > CD > IA > RA, respectively. The result of GIS mapping of the spatial Hg distribution suggested that the emissions of coal-power plant and coking plant were the main sources of THg in the dust, whereas the contributions of MSW landfills and iron and steel smelting related industries were not significant. However, the emission of MSW landfills was considered to be an important source of MeHg in the studied area. The result of health risk assessment indicated there was a high adverse health effect of the kindergarten dust on the children living in the educational area. Our study demonstrated that Hg in the dust samples could effectively reveal the atmospheric Hg deposition from different sources and the ecological and health effects of the enriched Hg in the studied city need further detailed investigations.

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