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ORIGINAL PAPER

Heavy metals and polycyclic aromatic hydrocarbons in municipal sewage sludge from a river in highly urbanized metropolitan area in Hanoi, Vietnam: levels, accumulation pattern and assessment of land application

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Abstract Concentrations of heavy metals and polycyclic aromatic hydrocarbons (PAHs) in sludge from Kim Nguu River, Hanoi, Vietnam, were analyzed to understand the contamination levels, distribution and accumulation pattern of municipal sludge from a highly urbanized area that receive direct discharge of wastewater. High concentrations of heavy metals such as As, Cd, Pb, Cu and Zn were observed in sludge, which were exceeded the Vietnamese regulation threshold values. In general, contamination status of heavy metals in sludge was in the similar range or slightly lower than those previously reported in sludge from the same area. The mean concentrations of As, Cd, Cr, Cu, Ni, Pb and Zn were 24.3, 2.65, 105, 166, 60.8, 73.7 and 569 mg/kg dry wt., respectively. Our result also indicates increased levels of PAHs, which are among the first data on PAHs accumulation in municipal sludge from metropolitan area in Vietnam. PAH concentrations ranged from 218 to 751 mg/kg dry wt. (mean: 456 mg/kg dry wt.), which were greater than those reported in sewage sludge from other countries as well as in sediments and soils collected from the same area. Accumulation pattern revealed the predominant of higher-ringed PAH compounds. Indicator ratios suggest the sources of PAHs were probably derived from biomass (wood and coal) and fossil fuel combustion and petroleum emissions. Most of the sludge samples contain PAHs concentrations exceeding various international guidelines values for sludge and sediment, such as probable effect levels, suggesting the possible risk for adverse biological effects in the study area and in the landfill sites where dredged sludge was dumped.

Keywords Kim Nguu River · Municipal sludge · Heavy metals · Polycyclic aromatic hydrocarbons

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Introduction

Vietnam has two biggest deltas: Red River Delta in the north and Mekong River Delta in the south. These two deltas inhabited by more than 30 million people, which are among the most densely populated areas in Asia–Pacific region. Hanoi is the capital city of Vietnam and located at the center of the Bac Bo Plain. Population of Hanoi reaches nearly 7.1 million in 2012, which increases almost three times since mid 1990s. Hanoi experienced rapid economic growth in recent years with uncontrolled environmental pollution due to various man-made chemicals. To Lich and

Kim Nguu River flows through major Metropolitan of Hanoi, which receive wastewater discharge of human and industrial activities of the city. The daily discharge volume of wastewater in the urban area of Hanoi is estimated to be 458,000 m^3 including municipal, medical and industrial sources. Since the wastewater treatment plant facilities are limited, the amount of sewage sludge generated from the wastewater treatment plant is small. The major amount comes mainly from excavated sludge of drainage system. In Hanoi, municipal sludge is collected and gathered to the city's landfill in Tu Hiep commune, Thanh Tri district. Sludge is only treated by dumping in landfill areas, which may cause serious environmental pollution as sewage sludge contains various kinds of contaminants as a results of direct discharge wastewater from Hanoi Metropolitan area.

The municipal sewage sludge can contain up to 300 different organic compounds, inorganic compounds and harmful microorganisms pathogens. The most frequently detected contaminants are polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), phthalic acid esters (PAEs), polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs), organochlorinated pesticides, chlorobenzenes (CBs), amines, nitrosamines, phenols (Perez et al. 2001; Busetti et al. 2006; Cai et al. 2007; Yan et al. 2009). The accumulation of organic compounds and heavy metals in sludge could cause difficulties for the use of municipal sewage sludge for agricultural application (Villar et al. 2006; Oleszczuk 2007; Khadhar et al. 2010). Furthermore, if sludge contains pollutants with increased concentrations, it is required to subject to an appropriate management and treatment. Regulations for threshold limit values of some potential toxic heavy metals in different kinds of soils were promulgated by Vietnamese Ministry of Natural Resources and Environment in 2008 (QCVN: 03/2008/BTNMT). However, regulations for polycyclic aromatic hydrocarbon (PAHs) are currently not available. While there are several studies on heavy metals contamination in the environment around Kim Nguu River and Hanoi Metropolitan area, there have been almost no studies on the occurrence of PAHs in these areas.

In Japan, up to 30 % of sewage sludge has been treated by biological methods toward products such as fertilizer used in agriculture (Ito et al. 2000). In the future, the percentage of treated sewage sludge will be definitely higher. In Vietnam, there have been researches about municipal sewage sludge treatment methods, but mainly focus on septic tank sludge. Currently, the direct use of municipal sewage sludge for agricultural soil application is not feasible due to its high pollution level. Therefore, municipal sewage sludge in urban areas of Vietnam needs to be treated with an appropriate treatment method to reduce pollution level, to produce utilization product after treatment and with low treatment cost. To meet those requirements, comprehensive in-depth studies are needed.

In the present study, we analyzed municipal sludge samples from Kim Nguu River for heavy metals and 16 target PAH compounds in order to evaluate the occurrence and accumulation characteristic of these contaminants in sludge. This is a part of the research program to develop an appropriate anaerobic digestion method to stabilize the municipal sludge and verify the content, fate and transformation of heavy metals and PAHs in municipal sludge from a highly industrialized and densely populated area of Hanoi Metropolitan. This may serves as important basis to provide better options for environmental management of the municipal sludge in Vietnam.

Materials and methods

Sampling locations

Kim Nguu River flows through the densely populated area of Hanoi City. The wastewater from the area of 6 km^2 flows into this river, which amounts to 139,000 m^3/day, being one-third of the wastewater produced in the city (Nguyen et al. 2007). The main industry that discharges wastewater into the river is textile and mechanical engineering and many small private workshops.

In this study, five sampling sites were selected along the river. The map showing sampling locations is shown in Fig. 1 and Table 1. Sediment samples were collected from the sites along the most important part of the Kim Nguu River in order to evaluate the environmental contamination by PAHs and heavy metals derived from the industrial and human activities around this metropolitan area.

Sludge samples were taken by Ekman grab sampler. Three replicate samples were collected from each sampling site with total 15 samples. The sludge

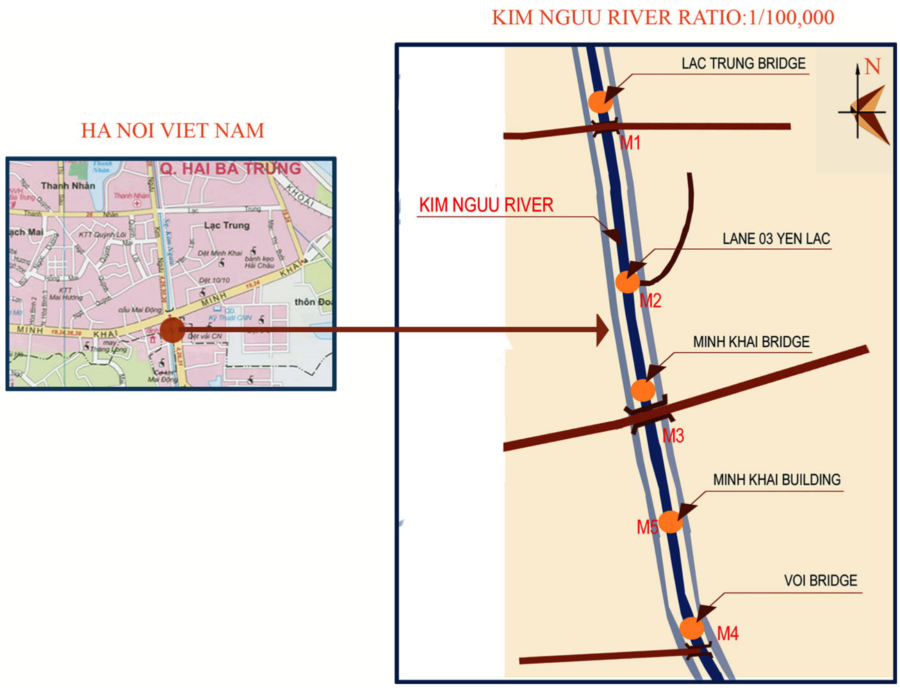

Fig. 1 *Map* showing the study areas in Kim Nguu River, Hanoi, Vietnam

Table 1 Information of sampling locations along the Kim Nguu River, Hanoi, Vietnam

Sampling points	Description	GPS position	
		Latitude	Longitude
M1	Lac Trung bridge	21°0′9.40″N	105°51′41.99″E
M2	Lane 03 Yen Lac	20°59′57.13″N	105°51′43.28″E
M3	Minh Khai bridge	20°59′47.54″N	105°51′44.32″E
M4	Voi bridge	20°59′28.59″N	105°51′46.41″E
M5	Minh Khai building	20°59′30.79″N	105°51′45.11″E

samples were immediately stored in a cool box until return to the laboratory where the sludge samples were stored at 4 °C for a maximum of one and a half days until further treatment. All sludge samples were collected at five locations from the top 20 cm of the sludge in the river.

Chemical reagents

Standard solutions of heavy metals are ICP single element standard solutions of 1,000 mg/L prepared in HNO_3 2–3 %, obtained from Merck Chemicals (Darmstadt, Germany). Nitric acid ultrapure 60 % and hydrogen peroxide (H_2O_2) were also obtained from Merck Chemicals. PAHs standards solutions were mixture of 16 PAHs with the concentration of 1,000 μg/mL prepared in toluene (PAH Mix 63-US EPA) provided by Dr. Ehrenstorfer Reference Materials (Augsburg, Germany). Anhydrous sodium sulfate was from Merck Chemicals.

Analysis of chemical and physical parameters of sludge samples

Chemical oxygen demand (COD) was determined by dichromate method and photometric measurement at wavelength of 605 nm; NH_4^+, NO_3^-, PO_4^{3-} were determined by UV–VIS spectrometric method; pH and electrical conductivity (EC) were measured by pH Cyberscan EuTech Con700 and Cyberscan 110; humidity was defined by drying at 105 °C during 24 h; volatile solid (VS) was determined by burning dry samples in ceramic cup at 550 °C during 8 h.

Chemical analysis of heavy metals

The sludge samples were completely dried at 40 °C during 24 h. Then, samples were grinded in a mortar and kept in a desiccator. The digestion procedure was applied following EPA Method 3050b. About 1 g of dry weight sample was added into each 200 mL Teflon vessel and amended 9 mL HNO_3 60 % and 1 mL H_2O_2 30 %. All sample vessels were setup on OMNI tray. Digestion was used MARS microwave. Two steps of microwave were setup such as step 1: temperature ramped to 165 °C, maximum pressure 350 psi, holding time 4 min; step 2: temperature ramped to 175 °C, maximum pressure 350 psi, holding time 20 min. After digesting and cooling, samples were filtered though filter paper. The volume of aliquot sample was filled up to 40 mL by de-ionized water. Heavy metals were determined by ICP OES (Thermo ICP OES, iCAP 6500) and calculated based on volume of aliquot sample and dry weight. Detection limits of the heavy metals are as follows: As: 1.1 mg/kg; Cd, Cr, Cu, Ni, Pb, Zn: 0.1 mg/kg dry wt.

Chemical analysis of PAHs

The sludge samples were preliminary dried at room temperature and in an oven at 40 °C until complete dry. Samples were grinded and sieved to remove large particles. The samples were extracted two times using 40 mL of dichloromethane in a tight Teflon tube under condition: temperature 80 °C, maximum pressure 350 psi and retention time 20 min (Barnabas et al. 1995). In total, 50 µL of d_{10}-phenanthrene solution (10-mg d_{10}-phenanthrene in 50 mL dichloromethane) was added to the sample as a surrogate standard before extraction. The supernatant was decanted after each cycle of extraction and then filtered through filter paper, sodium sulfate power added to remove excess water. The composite supernatants were evaporated using rotary vacuum evaporator at 40 °C, until sample volumes were smaller than 1 mL. The samples were fulfilled with dichloromethane to 2 mL, filtered through a PP-housing 0.45 µm syringe filter (Minisart® RC 15) and stored in glass vials sealed with Teflon-butyl rubber caps. Samples with complicated matrix interfere PAH signals; the extract was subjected to further clean up with silica gel column. The final extract was concentrated and injected into a gas chromatography with flame ionization detector (FID) for quantification.

Polycyclic aromatic hydrocarbons (PAHs) were analyzed using YL 6100 series gas chromatograph using Agilent J&W Advanced Capillary GC column HP-5 (30 m × 0.32 mm i.d.; film thickness 0.25 µm) and FID. A 2 µL aliquot of PAH sample was injected using an auto-sampler. Nitrogen was used as the carrier gas at a flow rate 2.5 mL/min. Inlet conditions are split ratio 5:1, split flow 10 mL/min, heater 200 °C and pressure 11.5 psi. The starting temperature was 150 °C, and the temperature was ramped to 190 °C at 8 °C/min with 5 min holding, ramped to 220 °C at 2 °C/min, ramped to 300 °C at 15 °C/min and then ramped to 310 °C at 2 °C/min with 2 min holding. Detector conditions were heater 300 °C, H_2 flow 36 mL/min, air flow 350 mL/min and make up flow 30 mL/min. Recoveries of the PAH compounds ranged from 80 to 96 %. Detection limit of the PAH compounds was ranged between 5.0 and 10 µg/kg dry wt.

Results and discussion

The physico-chemical indicators of sludge samples

The physico-chemical indicators of five sludge samples are stable. The pH values range from 7.04 to 7.41, CODt range from 79,900 to 83,030 mg/L, the VS is stable in the range from 19.2 to 23.5 % and other parameters were also not in large fluctuation among the samples analyzed (Table 2). The stability of the

Table 2 Physico-chemical parameters of sludge samples from Kim Nguu River, Hanoi, Vietnam

Samples	CODt (mg/L)	NH_4^+ (mg/L)	NO_3^- (mg/L)	PO_4^{3-} (mg/L)	EC (µS/cm)	TS (%)	VS (% TS)	pH
M1 ($n = 3$)	81,220	28.3	202	522	2,910	21.3	24.5	7.41
M2 ($n = 3$)	83,030	26.4	192	494	3,010	19.2	25.1	7.30
M3 ($n = 3$)	79,910	25.9	200	518	3,170	23.5	26.2	7.22
M4 ($n = 3$)	80,010	27.7	212	503	2,890	22.9	25.9	7.04
M5 ($n = 3$)	81,230	26.3	207	511	2,975	23.2	26.1	7.25

physico-chemical parameters suggests that sludge at Kim Nguu River has existed for relatively extended period of time.

Heavy metals in sludge samples

Concentrations of heavy metals in the sludge samples are shown in Table 3. Concentrations of As were ranged from 13.6 to 47.8 mg/kg dry wt. with an average level of 24.3 mg/kg dry wt. (Table 3). These levels were comparable with those reported in Kim Nguu and To Lich River reported in a recent survey (Marcussen et al. 2008). The highest concentrations were found in site M3 (mean: 47.8 mg/kg dry wt.), while concentrations in other sites are rather similar. According to (Marcussen et al. 2008), Kim Nguu and To Lich River sludge has been polluted with As due to wastewater discharge. Cadmium concentrations were also in the range to those reported in 2008 survey (Marcussen et al. 2008) with average concentration of 2.65 mg/kg dry wt. The levels of Cd in this study were still lower than those reported in a survey in 2005, showing increased Cd accumulation with the highest concentration up to 40 mg/kg dry wt. (Nguyen et al. 2007).

Average Cr and Cu concentrations were 105 and 166 mg/kg dry wt, respectively, which were somewhat lower than those reported in the previous survey (Marcussen et al. 2008). It should be noted the Cr concentrations in sewage sludge in this study were apparently lower than those reported in sediment and sludge from previous surveys (Marcussen et al. 2008; Nguyen et al. 2007). Lead concentrations were ranged from 24.4 to 220 with mean level of 73.7 mg/kg dry wt. These levels were clearly lower than those reported in these previous studies.

In general, our results indicate that heavy metal pollution in sludge from Kim Nguu River was generally lower than those reported previously, suggesting an improvement of the environmental quality in recent years. Several dredging campaigns of sediment and sludge from To Lich and Kim Nguu River have recently been implemented, and the quality of water and sediment may be improved. However, the degree of contamination is still high despite the declining trend. Most of the sludge samples contained As, Cd, Cu and Zn concentrations exceeding the Vietnamese regulation threshold levels (Table 4). In addition, differences in sampling sites, number of samples, changes in situation of waste discharge in recent years may account for the differences in heavy metals contamination between the two investigations.

Spatial distributions of heavy metals indicate increasing concentrations from site M1 to M3 and then decreasing (Table 3). Concentrations of heavy metals in the site M3 were the highest. Samples from this site receive direct discharge from the textile company and a number of mechanical workshops around this area. Nguyen et al. (2007) studied the impact of the industrial activity on the sediment quality in term of heavy metal pollution. The authors suggested different industrial sources around To Lich and Kim Nguu River metropolitan area for several heavy metals. In this study, heavy metal levels were found to be increased in the point receiving direct discharge from textile company and surrounding mechanical workshops. In general, concentrations from other sampling sites were relatively similar.

Kim Nguu River is one of the receiving wastewater rivers of Hanoi urban that still receives daily a large amount of wastewater from the operating industrial

Table 3 Concentration of heavy metals in the sludge samples from Kim Nguu River, Hanoi, Vietnam

Heavy metals	Concentration (mg/kg dry wt.)										
	M1 ($n = 3$)		M2 ($n = 3$)		M3 ($n = 3$)		M4 ($n = 3$)		M5 ($n = 3$)		Average
	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	
As	18.8–23.6	20.2	18.4–28.8	23.1	40.8–51.3	47.8	14.4–19.2	16.9	12.4–14.4	13.6	24.3
Cd	1.72–1.80	1.76	3.15–3.96	3.45	3.42–4.00	3.71	1.84–2.07	1.93	2.25–2.68	2.40	2.65
Cr	58.9–64.8	61.0	120–132	127	196–211	202	67.9–78.4	74.0	56.8–66.4	63.0	105
Cu	79.2–92.0	88.0	242–263	251	276–318	297	88.4–110	97.5	86.4–108	98.8	166
Ni	30.4–38.7	35.0	51.2–55.8	53.3	118–151	136	32.8–36.4	34.6	40.8–48.6	45.1	60.8
Pb	31.7–36.0	33.0	55.6–69.7	61.4	204–220	209	24.4–30.2	27.9	35.6–38.3	37.0	73.7
Zn	388–430	413	659–716	694	928–1,017	972	290–371	318	438–452	446	569

Table 4 Comparison of average concentration of heavy metals in the sludge samples in Kim Nguu River, Hanoi with Vietnam regulation threshold values

Heavy metals	Concentration (mg/kg dry wt.)					
	Average concentration in sludge samples	Agricultural soil	Forest soil	Residential soil	Commercial soil	Industrial soil
As	24.3	12 (100)[a]	12 (100)	12 (100)	12 (100)	12 (100)
Cd	2.65	2 (67)	2 (67)	5	5	10
Cu	105	50 (100)	50 (100)	70 (100)	70 (100)	100 (60)
Pb	73.7	70 (20)	100 (20)	120 (20)	200 (20)	300
Zn	569	200 (100)	200 (100)	200 (100)	300 (93)	300 (93)

[a] Values in parentheses indicate percentage of samples with heavy metals concentrations exceeding the Vietnamese regulation threshold values

factories in the city. This explains for the large difference in the concentration of heavy metals at different sampling points. For example, at M3 site (Minh Khai bridge), the concentration of some selected heavy metals is much higher than at other sampling points (Table 3). M3 is the focal point of many operating industrial factories such as Minh Khai Lock Factory and a number of textile companies.

The contamination pattern of heavy metals in the sewage sludge samples is usually in the order Cd < Pb < Cu < Zn (Chipasa. 2003; Pathak et al. 2009). Concentrations of heavy metals in this study were compared with those reported in other locations in the world to understand the magnitude of contamination (Table 6). Concentrations of Pb, Cu, Ni and Cr show similar contamination levels with those in sewage sludge in Beijing and Zhejiang, China (Dai et al. 2007; Hua et al. 2008). However, the average concentration of Zn in Kim Nguu sludge samples was 569 mg/kg dry wt. (Table 3), which is lower than the concentration of Zn from sewage sludge in Beijing (783–3,096 mg/kg dry wt.) and in sewage sludge from Zhejiang, China (1,406–3,699 mg/kg dry wt.). Concentrations of some heavy metals in sludge samples in Vietnam are lower than those in sewage sludge of wastewater treatment plants in China and Spain (Dai et al. 2007; Hua et al. 2008; Roig et al. 2012). However, the heavy metal content of sludge samples in this study was much higher than those in sediments at the sea port from Iran (Abdollahi et al. 2013) and higher than sediments samples from locations which are not receive direct discharge of untreated wastewater (Santschi et al. 2001).

The composition of sewage sludge is mainly due to characteristics of wastewater sources that enter the drainage system. In urban areas, heavy metals in sewage sludge mainly come from the sources such as human excretion products, cosmetic products, detergents and kitchen waste (Alloway and Jacson 1991). Heavy metals such as Cu, Cd, Pb, Hg and Cr are found at relatively high concentrations in sewage sludge (Xu et al. 2013) The total heavy metal content of sewage sludge is about 0.5–2.0 % (dry weight), and in some cases, may be as high as 4 % (wet weight), particularly for metals such as Cd, Cr, Cu, Pb, Ni and Zn (Xu et al. 2013). The accumulation of heavy metals in sludge is due to uptake of heavy metal ions from domestic and industrial wastewaters by microbial biomass of activated sludge (biosorption) and due to precipitation of sulfides in anaerobic digester. Chelating and subsequent intracellular accumulation of metals, adsorption onto the microbial cell walls, sheaths and capsules are the main mechanisms of biosorption (Chipasa 2003; Wang et al. 2005; Zhu et al. 2013).

Levels, accumulation pattern and sources of PAHs

Increased concentrations of 16 PAH compounds were found in sludge samples from Kim Nguu River, ranged from 218 to 751 mg/kg dry wt. (Table 5). Similar to the result of heavy metals, samples collected at site M3 contained the highest concentrations of PAHs (Minh Khai bridge). This site is the connection of many wastewater sources of small factories and mechanical workshops surrounding, and the vicinity of direct discharge of a textile company. Distribution of PAHs

Table 5 Concentrations of PAH compounds in the sludge samples from Kim Nguu River, Hanoi, Vietnam

PAHs (compounds)	Concentration (mg/kg dry wt.)										
	M1 ($n = 3$)		M2 ($n = 3$)		M3 ($n = 3$)		M4 ($n = 3$)		M5 ($n = 3$)		Average
	Range	Mean	Range	Mean	Range	mean	Range	mean	Range	Mean	
Naphthalene	1.65–2.14	1.95	1.15–1.27	1.22	1.26–1.60	1.42	0.58–0.78	0.69	1.05–1.29	1.20	1.29
Acenaphthylene	2.10–2.62	2.40	1.74–1.97	1.88	7.43–8.35	7.88	3.33–4.12	3.82	1.53–2.80	2.30	3.66
Acenaphthene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Fluorene	4.20–5.54	4.94	3.93–4.38	4.15	4.68–5.30	4.97	5.69–6.32	6.10	3.29–4.66	3.93	4.82
Phenanthrene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Anthracene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01–0.66	0.22	<0.01	<0.01	0.04
Fluoranthene	2.54–2.91	2.76	0.94–1.15	1.03	1.87–2.15	1.98	<0.01	<0.01	<0.01	<0.01	1.15
Pyrene	0.62–0.79	0.71	0.49–0.63	0.58	2.46–2.81	2.59	<0.01	<0.01	<0.01	<0.01	0.78
Benz[a]anthracene	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Chrysene	< 0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Benz[b]fluoranthene	0.61–0.74	0.64	0.62–0.73	0.69	1.57–1.76	1.68	1.92–2.13	2.05	0.90–1.15	1.03	1.22
Benzo[k]fluoranthene	4.50–5.03	4.72	3.31–3.61	3.42	7.85–8.12	7.97	2.33–2.79	2.60	2.19–2.89	2.55	4.25
Benzo[a]pyrene	12.8–14.3	13.6	11.5–12.2	11.9	6.07–6.66	6.40	9.71–12.2	10.9	6.90–7.59	7.22	10.0
Indeno[1,2,3-cd]pyrene	123–146	135	111–114	113	256–283	269	45.4–48.9	47.2	44.5–48.7	46.7	122
Dibenz[a,h]anthracene	96.1–120	107	85.5–93.1	89.9	187–214	201	54.9–63.9	59.1	44.0–46.5	44.0	100
Benzo[ghi]perylene	241–293	264	221–251	240	237–255	246	165–175	171	110–165	109	206
Total 2–4 rings PAHs	11.7- 13.7	12.8	8.54–9.10	8.86	18.3–19.2	18.8	9.60–11.8	10.8	5.87–8.71	7.42	11.7
Total 5–6 rings PAHs	478–580	525	435–472	458	696–759	732	279–302	293	201–221	211	444
Total PAHs	489–594	538	444–480	467	715–778	751	289–313	304	207–230	218	456

was rather similar to that of heavy metals, showing peak concentration in M3 site. In general, PAHs concentrations decreased toward downstream of the river. Information on the PAHs contamination in soils and sediment samples from metropolitan area in Vietnam is limited. Boll et al. (2008) investigated PAHs accumulation in soils and sediments from several locations in Hanoi city including Kim Nguu River. Spatial distribution of PAHs in sediment from Kim Nguu River was relatively similar to those observed in this study, showing a decrease in concentrations toward downstream of the river. However, total PAHs concentrations in the sludge from this study were apparently higher than those in soils and sediments collected in the same area (Boll et al. 2008). Kishida et al. (2007) reported PAHs contamination in different locations in metropolitan areas of Hanoi and Hochiminh City. The total PAHs concentrations ranged from 0.03 to 6.40 mg/kg dry wt. which were also much lower than those in sludge of the present study. Our result provides one of the first data on the PAHs contamination in sludge of river receiving non-treated wastewater from Vietnam.

Polycyclic aromatic hydrocarbons (PAHs) concentrations in sludge from Kim Nguu River in Hanoi, Vietnam, were compared with those from other locations to understand the magnitude of contamination (Table 6). Dai et al. (2007) reported PAHs concentrations in sewage sludge at six wastewater treatment plants in Beijing, China, ranged from 2.47 to 25.9 mg/kg dry wt. Similar research in Korea for sewage sludge at six treatment plants of domestic wastewater obtained total PAHs concentration in the range from 1.24 to 44.9 mg/kg dry wt. (Ju et al. 2009). PAHs levels in sludge samples of Kim Nguu River in Hanoi were much higher than total PAHs concentration of sewage sludge in China and South Korea. The reason is that sewage sludge samples were collected from Chinese and Korean wastewater treatment plants where sewage sludge is always transported and treated. The municipal sludge samples in Hanoi, Vietnam, were generally not subjected to treatment and usually mixed with long-term contaminated sediment in the river, leading to the increased PAHs concentrations. The occurrence of PAHs in sludges has been investigated since 1970s in industrialized countries such as USA, UK and Canada. The mean total PAHs were frequently found from 1 to 100 mg/kg dry wt., although values up to 2000 mg/kg dry wt. have also been reported (Cai et al. 2007). However, PAHs levels in sludge from Kim Nguu River which is river receiving non-treated wastewater in Hanoi are higher than those in sewage sludge and sediment from other locations in the world (Table 6).

Accumulation of PAHs in sludge samples revealed specific pattern, showing the predominant of higher-ringed PAH compounds, such as indeno[1,2,3-cd]pyrene, dibenz[a,h]anthracene and benzo[g,h,i]perylene (Table 5). Concentrations of potential toxic compound benzo[a]pyrene were also increased, accounting for relatively high proportion to total PAHs concentrations. In general, concentrations of 5–6-ringed PAHs were much higher than those of lower-ringed compounds (Table 5). Accumulation pattern of PAHs in sewage sludge often varied markedly depending on the complexity of the urban and industrial sources. PAH compounds' specific pattern of sewage sludge from sewage treatment plants in Spain were varied among different plants, showing higher proportion of higher-ringed PAHs in one plant and the predominant of 2–3 ringed compounds in the others (Perez et al. 2001). In wastewater treatment plants in Korea, the accumulation patterns were also different among plants, which could be due to the influence of diverse industrial sources (Ju et al. 2009). In China, high molecular weight compounds such as indeno[1,2,3-cd]pyrene and dibenzo[a,h]anthracene were also accounted for relatively larger proportions to total PAHs concentrations (Dai et al. 2007). In the present study, very high levels of high molecular weight PAHs were found, which is somewhat similar to that found in some wastewater treatment plants in China and Korea. The dominance of higher-ringed compounds suggests the pyrogenic origins of PAHs in this study.

To further access the PAHs accumulation characteristics in sewage sludge, the compound-specific patterns in samples from this study were compared with those reported in sediments and soils from Vietnam (Fig. 2). It is clear that the pattern in sewage sludge was different to the pattern observed in sediment and soil samples. Boll et al. (2008) examined accumulation pattern of PAHs in sediment and soil collected from Kim Nguu River with relatively similar areas of this study. Our results showed specific pattern of PAHs in sludge of river receiving non-treated wastewater, showing apparently higher proportion of 5- and 6-ringed compounds as compared to sediment

Table 6 Comparison of heavy metals and PAHs concentrations in sewage sludge and sediment from different locations

Location	Concentration (mg/kg dry wt.)								References
	Σ 16 PAHs	Heavy metals							
		As	Cd	Cr	Cu	Ni	Pb	Zn	
Vietnam									
Sludge, Kim Nguu River	456	24.3	2.65	105	166	60.8	73.7	569	This study
Sludge, Kim Nguu River (down stream)			3.5	585	475	151	665	520	Nguyen et al. (2007)
Sludge, Kim Nguu River		22–33	0.9–2.1	112–166	61.8–128	54–95	60.9–97.4	229–687	Marcussen et al. (2008)
Asia									
Sewage sludge, WWTPs, Beijing, China	2.47–25.9	21.2–26.0	5.9–13.0	45.8–78.4	131–394	49.3–95.5	57.5–109	783–3,096	Dai et al. (2007)
Sewage sludge, WWTPs, Zhejiang, China	33.7–87.5		2.1–23.4	22.2–453	210–1,191	25.1–102	41.2–452	1,406–3,699	Hua et al. (2008)
Sewage sludge, WWTPs, China	16								Cai et al. (2007)
Sewage sludge, WWTPs, Korea	1.24–44.9								Ju et al. (2009)
Sediment, Imam Khomeini Port, Persian Gulf, Iran	2.89–5.48				12.9–18.3	54–58.3	5.6–8.14	43.3–59.6	Abdollahi et al. (2013)
Europe									
Sewage sludge, WWTPs, Spain	1.13–5.52								Perez et al. (2001)
Sewage sludge, WWTPs, Spain			1.7–5.3	61.1–200	173–577	24.0–103	46.6–95.8	578–1,508	Roig et al. (2012)
Sludge, Sediment tank, WWTP, France	49.6–60.2								Huault et al. (2009)
America									
Sediment, Almendares River, Cuba			1.0–4.3	84.4–209	71.6–420		39.3–189	86.1–708	Rieumont et al. (2005)
Sediment, Mississippi River Delta, USA	0.66		0.18	71.6	21.3		27.4	144	Santschi et al. (2001)

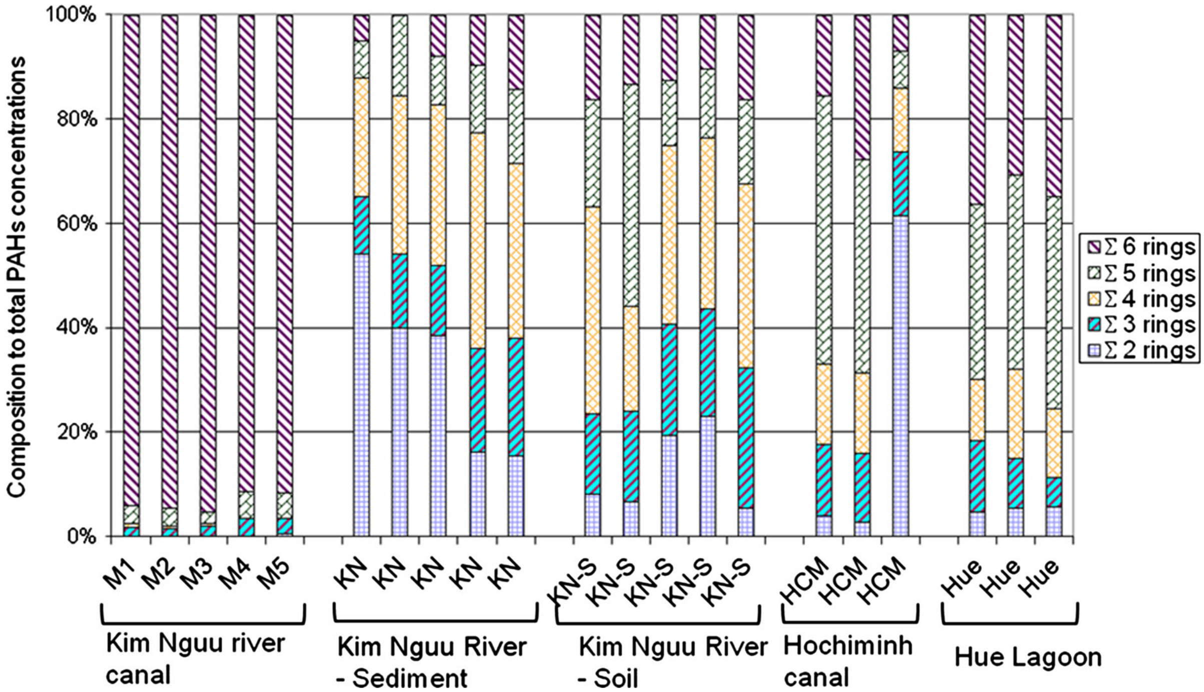

Fig. 2 Comparison of the PAH accumulation pattern of sewage, soils and sediments from Vietnam. *Source of data*: sediment and soil from Kim Nguu River: Boll et al. (2008); sediment from Hochiminh City and Hue: Kishida et al. (2007); and sewage sludge from Kim Nguu River: present study

and soil collected from the same river (Fig. 2). Sediment from highly industrialized and urbanized areas in Hochiminh City canals and lagoon from Hue also showed high proportion of higher-ringed PAH compounds (Kishida et al. 2007). The specific pattern with high concentrations of 5- and 6-ringed PAH compounds in this study indicates the strong influence of the pyrogenic sources from the Kim Nguu and the surrounding urbanized and industrial areas.

Polycyclic aromatic hydrocarbons (PAHs) are formed by combustion and incomplete thermal decomposition of many different organic substances such as coal, tobacco, fossil fuel, even cooking in the family (Gu et al. 2003; Shen et al. 2007; Nghiem and Cung. 2010). PAHs transfer through the atmosphere and accumulate in soil and water. PAHs exist on the surface of road through the wastewater by rain runoff leading to the accumulation in sewage sludge (Shen et al. 2007; Larsen et al. 2009).

Typically, waste-derived fossil fuels such as petroleum contains PAHs are mainly lower molecular weight or lower-ringed PAH compounds. The products of combustion and thermal decomposition contain PAHs with higher molecular weight (Dai et al. 2007; Shen et al. 2007; Sanctorum et al. 2011). Lower-ringed PAHs such as acenaphthene, phenanthrene, benz[a]anthracene and chrysene were not detected in sewage samples (Table 5). Anthracene was detected only in samples M4. Low molecular weight (LMW) to high molecular weight (HMW) ratios >1 indicate the dominance of petrogenic sources and LMW/HMW < 1 indicate pyrolytic sources. (Yan et al.2009; Abdollahi et al. 2013). The LMW/HMW values in sludge samples from all stations were less than 1, indicating that the PAHs in sludge in this region are derived mainly from pyrolytic sources. To further assess the potential source of PAHs in sludge samples in this study, several PAH indicator ratios were calculated and compared with those in some typical sources reported in the literature (Fig. 3). Ratio of fluoranthene/(fluoranthene + pyrene) between 0.4 and 0.5 indicates fossil fuel combustion. The ratio greater than 0.58 suggests biomass (wood) and coal combustion. A ratio of indeno[1,2,3-cd]pyrene (IP)/(IP + benzo[ghi]perylene) less than 1.8, between 0.2–0.5, and >0.5 suggests petroleum, fossil fuel and

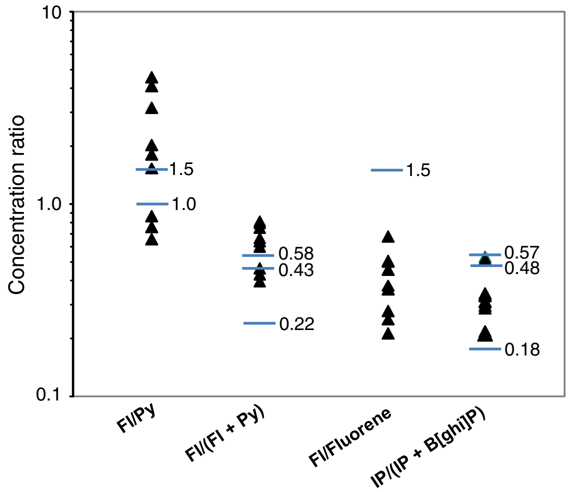

Fig. 3 PAH indicator ratios in sewage sludge from Kim Nguu River and potential sources reported in the literature. Ratio values indicating potential sources were cited from Kilemade et al. (2004), Donahue et al. (2006) and Yunker et al. (2002a, b). *Fl* fluoranthene; *Py* pyrene; *IP* indeno[1,2,3-cd]pyrene; *B[ghi]P* benzo[ghi]perylene

biomass (wood, coal) combustion, respectively (Yunker et al. 2002a, b). In the present study, Fl/(Fl + pyrene) in some samples were higher than 0.5, which may suggest biomass combustion sources. The ratio IP/(IP + benzo[ghi]perylene) was in the range of 0.18–0.57, indicating possible fossil fuel combustion origin. Overall, indicator ratios in sewage sludge from Kim Nguu River, Hanoi are likely derived from local sources due to fossil fuel and high temperature biomass combustion. In addition, several samples have Fl/(Fl + pyrene) ratio around 0.43 which may be related to the petroleum emission sources. Heavy traffic is also very common in the Metropolitan area, and Kim Nguu River is among the most densely populated areas in Hanoi city.

Assessment of agricultural land application

The application of these sludges in agriculture can be advantageous to the soil because they modify soil structure and provide organic matter and nutrients. In Europe, The Urban Waste Water Directive 91/271/EEC was amended by 98/15/EC came into force in 2005 and sets more stringent quality standards for wastewaters. The main article of the Urban Waste Water Treatment Directive dealing with sludge is Article 14, where it is declared that "*sludge arising from waste water treatment shall be re-used whenever appropriate*" (Fytili and Zabaniotou 2008). In Japan, up to 30 % of sewage sludge has been treated by biological methods toward products such as fertilizer used in agriculture (Ito et al. 2000). On the other hand, utilization as a fertilizer has some risks namely, the accumulation of pollutants in soils, plants, animal pastures and the subsequent entry into the food chain of toxic hazardous chemicals (Villar et al. 2006).

In order to assess the possibility of using sludge directly for land application needed to compare the levels of pollutants in sludge with regulation threshold levels. For heavy metals, most of the sludge samples contained As, Cd, Cu and Zn concentrations exceeding the Vietnamese regulation threshold levels (Table 4). Currently in Vietnam, there is no national regulation for PAH compounds in the soil and sediment. Sweden has regulation for six PAHs including fluoranthene, benzo[a]-pyrene, benzo[b] fluoranthene, benzo[k]fluoranthene, benzo [g,h,i]perylene and indeno[1,2,3-cd]pyrene) in sewage sludge that can be used for agricultural land with maximum concentration is 3 mg/kg dry wt. (Dai et al. 2007). European Union (EU) regulation value for total concentration of nine PAH compounds (acenaphthene, phenanthrene, fluorene, fluoranthene, pyrene, benzo[b + j+k]fluoranthene, benzo[a]pyrene, benzo[g,h,i]perylene and indeno[1,2,3-cd]pyrene) in sewage sludge for agricultural land use is 6 mg/kg dry wt. (Ju et al. 2009). Further, some other countries also prescribed not allowed concentration higher than 1 mg/kg dry wt. for benzo[a]pyrene (Villar et al. 2006). In the present study, concentrations of benzo[a]pyrene alone in the sludge samples from Hanoi metropolitan area (Table 5) exceeded the regulation values for EU and Sweden. Thus, it is not feasible to use directly sludge for agricultural land application. Therefore, it is essential that regulation threshold levels for PAHs in the agricultural soils and sediment should be developed in Vietnam. In addition, treatment technology to minimize residues of contaminants in sludge also needed to further develop strategies of utilizing sludge for agricultural land application. Given the low organic matter content and high concentration of contaminant in sewage sludge in Kim Nguu River, aerobic treatment processes based on the activity of *Thiobacillus ferrooxidans* bacteria can be a potential technology for the removal of heavy metals and enhance the biodegradation of PAHs.

Potential toxicological significance

Increased PAHs concentrations were found in sludge samples in Kim Nguu River suggest potential toxic implications for environmental quality and human health. The magnitude of PAHs contamination was considered in comparison with several international guideline values.

The probable effect levels (PELs) for adverse biological effects (CCME 2002) in freshwater sediment can be used to estimate the possible biological effects of PAHs in sewage sludge. The PEL is a level above which adverse effects are likely to occur. MacDonald et al. (2000) also developed a consensus for various guideline values including the probable effect concentration (PEC). To evaluate the toxicological relevance of the PAHs contamination sewage, their spatial distribution was considered in comparison with the Canadian PELs and the consensus-based PECs proposed by MacDonald et al. (2000) (Fig. 4).

Most of the PAH compounds have concentrations in sludge samples in Kim Nguu River, Hanoi exceeded the probable effect-based guideline values (Fig. 4). Only concentrations of fluoranthene and pyrene were lower than the guideline values in few samples. Increased PAH levels in sewage sludge could be of concern and may have potential to cause adverse biological effects. In Hanoi, municipal sludge dredging from the rivers is often dumped in landfill sites. Increased PAHs contamination in sludge may affect the local environment, and more importantly, may increase the degree of human exposure to these compounds. Further systematic investigations to monitor the extent and temporal trend of PAHs and other persistent toxic substances in sewage sludge and sludge from Kim Nguu River as well as other urbanized and industrialized areas in Hanoi City are, therefore, needed to help provide appropriate countermeasures for an effective treatment technology.

Conclusions

The purpose of this study was to determine the concentration of heavy metals and PAHs in the Kim Nguu River sludge for the purpose of prioritizing future municipal sludge treatment efforts. Our result is among the first data on PAHs accumulation in sludge

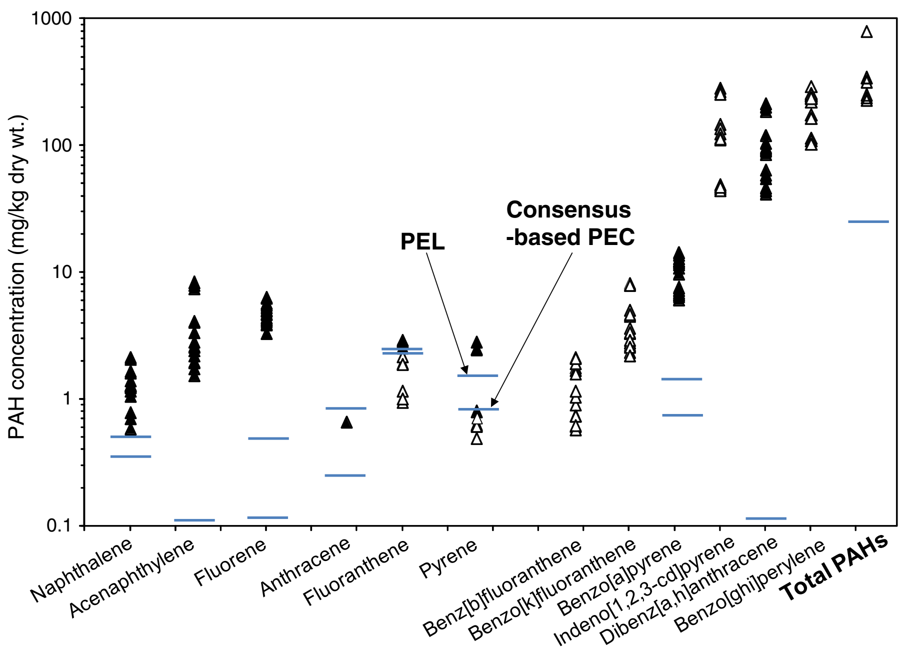

Fig. 4 PAH concentrations in sewage sludge samples in Kim Nguu River in comparison with guidelines values. *Blue lines* indicate the guideline values for different PAH compounds. *PEL* probable effect level for adverse biological effect proposed by Canada (CCME 2002). *Consensus-based PEC* consensus-based probable effect concentrations proposed by MacDonald et al. (2000)

from metropolitan area in Vietnam. Increased accumulation of heavy metals and PAHs in the sludge samples from Kim Nguu River in Hanoi, Vietnam, was observed. Most of the samples contained heavy metals concentrations (for As, Cd, Pb, Cu and Zn) and PAH compounds exceeding the Vietnamese and International standard values, suggesting possible risk for toxic implications on ecosystem and human exposure in the study area as well as the landfill sites where municipal sewage sludge were dumped. Further comprehensive investigations on the fate and distribution of potential toxic contaminants such as pesticides, chlorophenols, dioxins, furans and dioxin-like PCBs are important to provide a better baseline information of anthropogenic environmental pollutants in highly urbanized and industrial areas in Vietnam.

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