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Heavy Metals in Sediments and Water at the Chao Phraya River Mouth, Thailand

Varinporn Asokbunyarat* **and Sanya Sirivithayapakorn**
Department of Environmental Engineering, Faculty of Engineering,
Kasetsart University, Bangkok 10900, Thailand
*Email : varinporn.a@ku.th

Abstract

Sediment and water samples collected from 20 sampling stations located in the Chao Phraya River Mouth, Thailand during 2017 and 2019 were analyzed for concentrations of mercury (Hg) as well as other heavy metals including cadmium (Cd), zinc (Zn), nickel (Ni) and lead (Pb). The results indicated that the enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in sediments of the Chao Phraya River Mouth during 2017 and 2019. The higher concentrations of heavy metals were found in the suspended sediment in comparison to the bottom sediment. The concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) in water of the Chao Phraya River Mouth during 2017 and 2019 were lower than the coastal water quality standard in Thailand. Because of the high freshwater inflow and the discharge of low amount of heavy metals in the treated industrial effluent to the Chao Phraya River, the high levels of heavy metals in sediments of the Chao Phraya River Mouth might be caused by weathering of rock, sediment transport and deposition within the Chao Phraya River Basin. The spatial variation of heavy metals in sediments of the Chao Phraya River was existed, due to adsorption and co-precipitation of dissolved heavy metals onto sediments and aggregation-sedimentation of suspended sediment during transport. The seasonal variation of dissolved heavy metals in water of the Chao Phraya River Mouth was observed, due to accumulation and release of dissolved heavy metals from sediments.

Keywords : Heavy metals; Source identification; Water; Sediment; Suspended sediment; the Chao Phraya River Mouth

Introduction

In Thailand, the Chao Phraya River, which is the largest and most important river, passes through 10 cities, including Nakhon Sawan, Uthai Thani, Chai Nat, Singburi, Aug Thong, Ayutthaya, Pathum Thani, Nonthaburi, Bangkok, and Samut Prakan. In the Chao Phraya River Basin, many developments with high rate of urbanization and industrialization along the river banks and south of the Bangkok Metropolis, as well as agricultural modernization along the northern section of the river have been proceeding continuously over the last decade. Such developments possibly discharge wastewater or non-standard effluent to the river, thereby deteriorating aquatic environment and natural resources [1, 2]. Wastes of agrochemicals, urban areas and numerous industries, such as electroplating and electronic equipment, located along the river and large canals connecting the river, either partially treated or without treatment, are discharged into the Chao Phraya River, the Chao Phraya River Mouth and eventually into the Upper Gulf of Thailand causing water quality deterioration with pollutants, including heavy metals [3].

In the Chao Phraya River Mouth, it has been faced the problem from heavy metals. A survey by Polprasert, 1982 [1] found that the accumulation of Cd and Pb in water of the Chao Phraya River Mouth was significant. Wijaya et al., 2013 [4] indicated that the high concentrations of heavy metals (Cd, Cu, Cr, Pb and Zn) in sediment of the Chao Phraya River were observed, reflecting primarily lithogenic baseline concentration of heavy metals in unpolluted sediment. Qiao et al., 2015 [5] found that the concentrations of heavy metals (Cu, Pb, Zn, Cr and Cd) in sediment of the Chao Phraya River were higher than that of the Bang Pakong River, the Tha Chin River and the Mae Klong River. Mclaren et al., 2004 [3] reported that temporal variation of cadmium concentration in water and sediment of the Chao Phraya River was observed, most probably related to seasonal high water levels and sediment flushing.

In the aquatic environment, contamination of heavy metals is a serious threat due to its abundance, accumulation, persistence and environmental toxicity. Both natural actions and anthropogenic activities are responsible for the abundant of heavy metals in the aquatic environment. The heavy metals can be accumulated and magnified to mussels, oysters, shrimps and fish, and can be transferred to humans with the food chain pathways. The increasing pollutions by heavy metals have a significant adverse health effects for invertebrates, fish and humans [6-8].

The principal comportment of metals is a function of the water chemistry and sediment in the natural water body. During transportation of heavy metals in the riverine system, it might undergo frequently changes due to dissolution, precipitation and sorption phenomena [6]. The heavy metal concentrations in sediment are extremely higher than that in water because heavy metals tend to accumulate in bottom deposits [7]. However, heavy metals amassed in bottom sediment are more likely to be re-suspended and cause secondary contamination of heavy metals to the water column [8].

The aims of this study were (i) to monitor heavy metals (Hg, Cd, Pb, Ni and Zn) in sediments and water of the Chao Phraya River Mouth, Thailand during 2017 and 2019, (ii) to assess contamination levels of heavy metals, seasonal and spatial variation of heavy metals in sediments and water of the Chao Phraya River Mouth and (iii) to identify possible sources of heavy metals in sediments and water of the Chao Phraya River Mouth.

Methodology

Study area and sampling

Sediment and water samples were collected from 20 sample locations of the Chao Phraya River Mouth (**Figure 1**). Sampling was performed in four phases: firstly, January 2017 (dry season); secondly, June 2017 (wet season); thirdly, July 2018 (wet season) and fourthly, January 2019 (dry season).

A total of 160 samples (80 sediment samples and 80 water samples) were collected. About 100 g of surface sediment samples (15 cm depth) and 1 L of surface water samples were collected from 20 sampling stations of the Chao Phraya River Mouth, using a sediment collector and a grab method, respectively during the lowest tide. After collection, sediment and water samples were acidified with nitric acid (pH<2), stored at 4°C in the dark and transferred to the laboratory.

Sample digestion and analysis of samples

The sediment, and filtered and non-filtered water samples were digested with HNO_3-HF-HCl acid. 1 g of each sample was weighted into microwave vessels and digested with 3 mL 65% HNO_3, 1 mL 40% HF and 0.8 mL 37% HCL. The digestion was carried out using a microwave digester for 26 min. The digests were cooled to room temperature, filtered through a 0.2 μm membrane filter and diluted to 20 mL with distilled water. The filtrate from sediment samples was also analyzed for total heavy metals (Hg, Cd, Pb, Ni and Zn) using ICP-OES. The filtrate from filtered and non-filtered water samples was analyzed for dissolved and total heavy metals (Hg, Cd, Pb, Ni and Zn), respectively using ICP-OES. Difference of total and dissolved heavy metals in water was reported as values of heavy metals in suspended sediment.

Results and Discussion

Heavy metal concentration in bottom sediment

The mean values of heavy metals in sediment monitored at the Chao Phraya River Mouth, Thailand on four times during 2017 and 2019 (January 2017; June 2017; July 2018; January 2019) are summarized in **Figure 2**. The results indicated that the enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in sediment of the Chao Phraya River Mouth during 2017 and 2019. The concentrations of lead (Pb), zinc (Zn) and nickel (Ni) in sediment monitored in January 2017 were similar to those concentrations in sediment

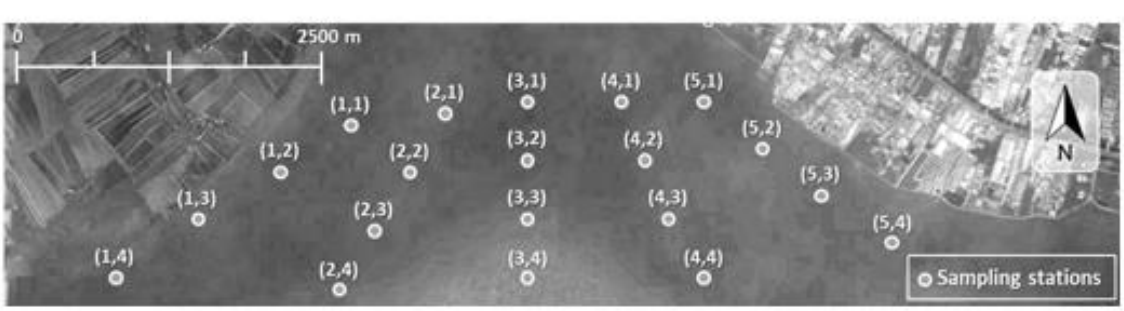

Figure 1 Sampling stations of sediment and water at the Chao Phraya River Mouth

monitored in July 2018 and January 2019. The mean concentrations of lead (Pb), zinc (Zn) and nickel (Ni) in sediment monitored in January 2017, July 2018 and January 2019 were 0.010-0.014 mg/kg for Pb, 0.017-0.022 mg/kg for Zn and 0.002-0.006 mg/kg for Ni. The lowest concentrations of cadmium, Cd (0.0003-0.0005 mg/kg) were accumulated in sediment monitored during 2017 and 2019. The mean concentrations of heavy metals in sediment monitored during 2017 and 2019 decreased on the following order: Zn > Pb > Ni > Cd. On the other hand, the mean concentrations of mercury (Hg) in sediment decreased significantly with increasing periods of monitoring. According to Menasveta and Cheevaparanapiwat, 1981 [9] and Hungspreugs and Yuangthong, 1983 [10], the high concentrations of lead (Pb) and mercury (Hg) were found in bottom sediment and green mussel of the Chao Phraya River Mouth.

Figure 2 indicates that the concentrations of mercury (Hg), zinc (Zn), and lead (Pb) in sediment of the Chao Phraya River Mouth in June 2017 and July 2018 in wet season were lower than those concentrations in January 2017 and January 2019 in dry season. According to Mclaren et al., 2004 [3], cadmium concentrations in sediment of the Chao Phraya River in wet season were lower than those concentrations in dry season. This might be due to flushing of sediment by large volumes of rapid moving water during wet season. On the other hand, Sirirattanachai and Utoomprurkporn, 2005 [11] found that mercury concentrations in sediment collected from 94 km upstream to the Chao Phraya River Mouth in wet season were higher than those concentrations in dry season, possibly due to re-suspension, suspended sediment transport and deposition.

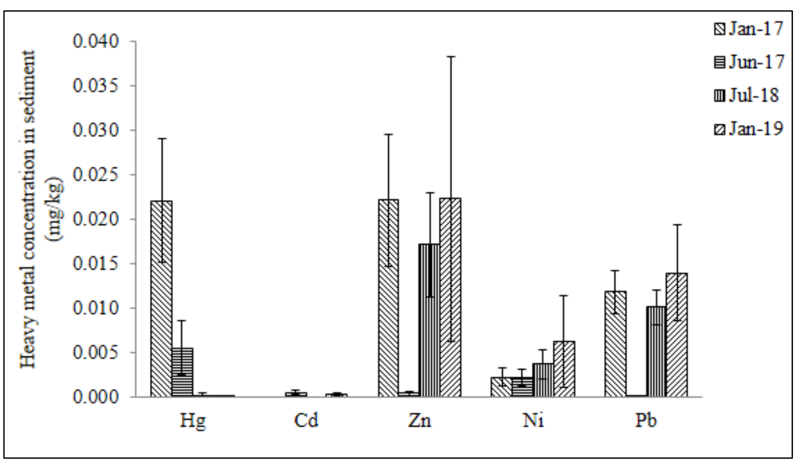

Figure 2 Mean values of heavy metals in sediment in the Chao Phraya River Mouth during 2017 and 2019. Each error bar indicates the standard deviation of sediment samples collected from 20 sample locations of the Chao Phraya River Mouth.

The concentrations of heavy metals in sediment of the Chao Phraya River Mouth, Thailand with other World rivers and river mouths are presented in **Table 1**. The results showed that the values of heavy metals (Hg, Cd, Zn, Ni and Pb) in sediment monitored at the Chao Phraya River Mouth, Thailand during 2017 and 2019 were detected in very low concentrations compared to sediment of other World rivers and river mouths. The seasonal and spatial variations of heavy metals in sediment of the Chao Phraya River, Thailand, the Liuyang River, China, the Karnaphuli River, Bangladesh and the Ghaghara River, India and in sediment of the Chao Phraya River Mouth, Thailand and the Yangtze River Mouth, China were significant [4, 6, 12-14]. Wijaya et al., 2013 [4] reported that river sediments collected from 72 km upstream to 27 km downstream from the Chao Phraya River Mouth showed spatial variation of heavy metals (Cd, Cu, Cr, Pb and Zn), possibly due to adsorption and co-precipitation of dissolved heavy metals onto suspended and bottom sediments, and aggregation-sedimentation of suspended sediment during transport.

The suspended sediment in the river might be caused by weathering of rock through water and wind which is continuously transported by river flow, tide and seasonal monsoon, and deposited along the river flow slow down, especially at the inside of river bends, and river mouths [5]. Because of the high freshwater inflow in the Chao Phraya River and the discharge of low amount of heavy metals contained in the treated industrial effluent into the Chao Phraya River, possible sources of heavy metals in sediment of the Chao Phraya River were not anthropogenic, but rather lithogenic. Wijaya et al., 2013 reported [4] that the values of heavy metals detected in sediment of the Chao Phraya River represented mainly lithogenic baseline concentration of heavy metals in the unpolluted sediment.

Heavy metal concentration in suspended sediment

The mean values of heavy metals in suspended sediment monitored at the Chao Phraya River Mouth in July 2018 and January 2019 are shown in **Figure 3**. The results indicated that the enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in suspended sediment of the Chao Phraya River Mouth during periods of monitoring. The mean concentrations of heavy metals in suspended sediment of the Chao Phraya River Mouth in July 2018 and January 2019 decreased on the following order: Pb > Ni > Zn > Cd > Hg. The high contamination levels of heavy metals in the river mouth suspended sediment might be caused by weathering of rock through water and wind which are natural sources of heavy metals in the environment, and the suspended sediment transport to the river mouth by river flow, tide and seasonal monsoon [15, 16]. Besides natural origin, the variation of heavy metals in the river mouth suspended sediment may be due to adsorption and co-precipitation of dissolved heavy metals onto the suspended sediment.

Table 1 Heavy metals in sediment of the Chao Phraya River Mouth, Thailand and other World rivers and river mouths

Metals	Chao Phraya River Mouth Sediment, Thailand	Chao Phraya River Sediment, Thailand		Liuyang River Sediment, China	Yangtze River Mouth Sediment, China	Karnaphuli River Sediment, Bangladesh	Ghaghara River Sediment, India
		72 km	27 km				
Hg, mg/kg	0.0001-0.022				0.065		
Cd, mg/kg	0.0003-0.0005	0.13	0.22	1.24	0.130	0.63-3.56	0.21-0.28
Zn, mg/kg	0.0004-0.022	21.9	103	138.48	71.5		13.26-17.59
Ni, mg/kg	0.002-0.006			17.48			15.29-25.59
Pb, mg/kg	0.0001-0.014	15.1	28.8	37.82	25.8	21.98-73.42	10.71-14.26
Reference	This study	[4]		[13]	[12]	[6]	[14]

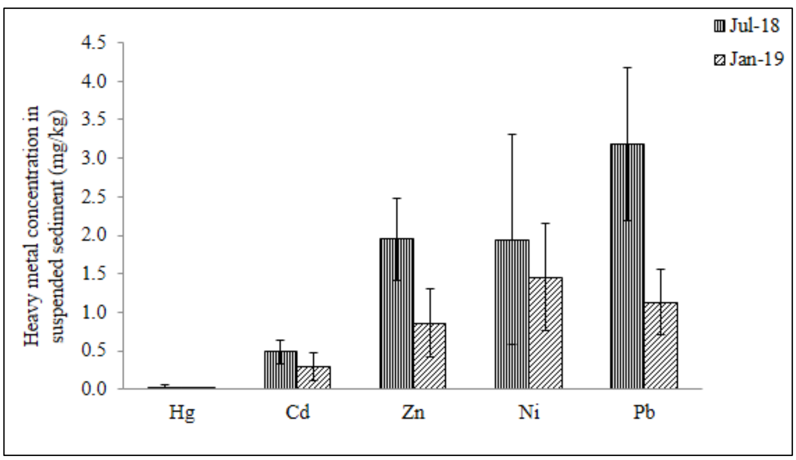

Figure 3 Mean values of heavy metals in suspended sediment in the Chao Phraya River Mouth during 2018 and 2019. Each error bar indicates the standard deviation of suspended sediment samples collected from 20 sample locations of the Chao Phraya River Mouth.

Figure 3 shows that the mean concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) in suspended sediment of the Chao Phraya River Mouth in July 2018 in wet season were higher than those concentrations in January 2019 in dry season. The seasonal variation of heavy metals in the river mouth suspended sediment might be due to the fast river flow in wet season, result in more suspended sediment transported to the river mouth. Furthermore, Figure 2 and Figure 3 indicate that the mean concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) in the river mouth suspended sediment were higher than those concentrations in the river mouth bottom sediment. The results of this study were similar to the results of Sabri et al., 1993 [17] and Asokbunyarat and Sirivithayapakorn, 2017 [18]. The low contamination levels of heavy metals in the river mouth bottom sediment might be due to dilution with organic matter and other pollutants.

Heavy metal concentration in water

The mean values of dissolved heavy metals in water monitored at the Chao Phraya River Mouth on four times during 2017 and 2019 (January 2017; June 2017; July 2018; January 2019) are presented in Figure 4. The mean values of dissolved heavy metals in water monitored in January 2017, which decreased on the following order: Pb > Zn > Ni > Cd > Hg, were higher than that of heavy metals monitored in June 2017, July 2018 and January 2019. However, in comparison with the monitoring data during June 2017 and January 2019, the mean values of dissolved heavy metals in water decreased continuously. The mean values of dissolved heavy metals in water monitored during June 2017 and January 2019 decreased on the following order: Zn ≥ Cd ≥ Pb ≥ Ni > Hg.

The mean values of dissolved mercury (Hg) monitored in water during January 2017 and January 2019 were in the range of 0.001 to 0.008 µg/L which were detected at very low concentrations compared to other dissolved heavy metals. The mean values of dissolved cadmium (Cd) and lead (Pb) in water during January 2017 and January 2019 were detected at higher concentrations than dissolved mercury (Hg). The mean values of dissolved lead (Pb) in water in January 2017 were detected at very high concentrations compared to the dissolved cadmium (Cd) and other dissolved heavy metals. However, the mean values of dissolved lead (Pb) in water during June 2017 and January 2019 were detected at lower concentrations than in January 2017. During June 2017 and January 2019, the mean values of dissolved lead (0.002-0.157 µg/L) in water were similar to the dissolved cadmium (0.001-0.192 µg/L), zinc (0.002-0.201 µg/L) and nickel (0.003-0.052 µg/L). The results of this study were similar to the results of Polprasert, 1982 [1] who found that accumulation of cadmium (Cd) and lead (Pb) in water at the Chao Phraya River Mouth was significant.

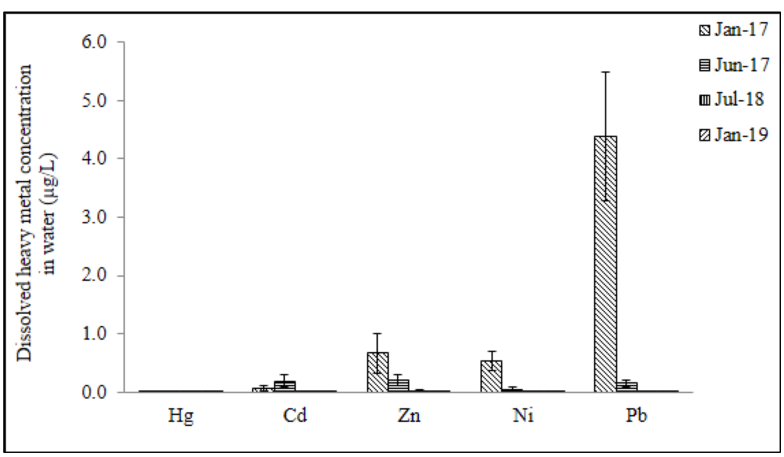

Figure 4 Mean values of dissolved heavy metals in water in the Chao Phraya River Mouth during 2017 and 2019. Each error bar indicates the standard deviation of water samples collected from 20 sample locations of the Chao Phraya River Mouth.

The variation of dissolved heavy metals in the river mouth water might be due to accumulation and release of dissolved heavy metals from the river suspended and bottom sediments. During transportation of heavy metals in the riverine system, it might undergo frequently changes due to dissolution, precipitation and sorption phenomena [6, 8]. According to Huang et al., 2017 [19], heavy metals (Cr, Ni, Cu, Zn, Cd and Pb) in sediment of the Huangpu River, China had a low release flux, among them Cu and Pb had the highest release flux, and dissolved Cu and Pb were fluctuating during the experimental period.

Some heavy metals dissolved in the river water might be attributed from the anthropogenic origins which include discharge of heavy metal-contaminated industrial effluent, either treated or without treatment. However, the heavy metal-contaminated wastewater of factories in the industrial zone, park and estate in cities located along the Chao Phraya River, including Ayutthaya, Phatumthani, Bangkok and Samutprakan is treated before discharge of industrial effluent into canals and rivers, regarding law-established industrial effluent standard, result in the discharge of low amount of heavy metals contained in industrial effluent into the water bodies. According to Mingkhwan and Worakhunpiset, 2018 [20], the levels of heavy metals (Cd, Cr, Cu, Mn, Ni, Pb and Zn) in the surface water collected near the industrial estate in Uthai district and Bangpa-in district, Ayutthaya, as the water source receiving industrial effluent did not exceed the permissible surface water quality standard in Thailand.

Figure 4 shows that the concentrations of heavy metals in water of the Chao Phraya River Mouth in June 2017 and July 2018 in wet season were higher than those concentrations in January 2019 in dry season. This might be due to a period of high rainfall and subsequent fast river flow involving movement of suspended sediment and release of more dissolved heavy metals in the waterway system. According to Mclaren et al., 2004 [3], in August and September (wet season) the change occurred giving rise to the increased cadmium concentrations in water of the Chao Phraya River.

The mean values of all dissolved and total heavy metals (Hg, Cd, Zn, Ni and Pb) in water monitored at the Chao Phraya River Mouth during 2017 and 2019, as shown in Table 2, were lower than the coastal water quality standard in Thailand. The heavy metals in water monitored at the mouths of other World rivers are also shown in **Table 2**. According to Chaiyara et al., 2013 [21], the concentrations of Cd, Zn and Pb in water monitored at the mouths of the Chao Phraya River, the Tha Chin River and the Mae Klong River, Thailand in 2013 were lower than those heavy metals in water monitored at the Chao Phraya River Mouth, Thailand during 2017 and 2019. Compared with the heavy metals in water in the coastal area of other countries, the concentrations of Pb in water monitored at the Chao Phraya River Mouth, Thailand were similar to lead (Pb) in water monitored at the coastal area of Tuaran, Sabah, Malaysia and the Yangtze River Estuary, China [12, 16]. And, the concentrations of Hg and Zn in water monitored at the Yangtze River Estuary, China were higher than those concentrations in water monitored at the Chao Phraya River Mouth, Thailand. In the Odiel River Mouth, Spain, the concentrations of heavy metals in water were very high. The heavy metal-contaminated water in the Odiel River Mouth was caused by acid mine drainage from the abandoned mines and waste in mine dumps [22].

Table 2 Heavy metals in water monitored in the mouths of the Chao Phraya River, Thailand and other World rivers

Metals	Chao Phraya River Mouth Water, Thailand		Chao Phraya River Mouth Water, Thailand	Tha Chin River Mouth Water, Thailand	Mae Klong River Mouth Water, Thailand	Odiel River Mouth Water, Spain	Water of Coastal area of Tuaran, Sabah, Malaysia	Yangtze River Estuary Water, China	Coastal water quality standard in Thailand
	DHM	THM							
Hg, μg/L	0.001-0.008	0.001-0.007						0.036	0.1
Cd, μg/L	0.001-0.192	0.029-0.193	0.010-0.030	0.010-0.030	0.020	100	0.337	0.041	5
Zn, μg/L	0.002-0.668	0.102-1.047	0.150-0.190	0.360-0.530	0.160-0.190		0.822	8.910	50
Ni, μg/L	0.003-0.540	0.052-0.541				300			-
Pb, μg/L	0.002-4.392	0.190-4.428	0.330-0.430	0.150-0.430	0.080	240	5.560	0.829	8.5
Reference	This study		[21]			[22]	[16]	[12]	PCD

DHM: Dissolved heavy metals, THM: Total heavy metals

Inflow of freshwater and input of total heavy metals to the Chao Phraya River Mouth during 2017 and 2019 are shown in **Table 3**. The results indicated that input of total heavy metals to the Chao Phraya River Mouth varied with inflow of freshwater, except in January 2017. In January 2017 (dry season), the Chao Phraya River Mouth received low freshwater inflows, 9.1×10^6 m^3/d, but the contamination of heavy metals entering the river mouth was concentrated – the load per unity of volume, 6.097 μg/L. Consequently, the input of total heavy metals to the river mouth was very high, 55.483 kg/d. In June 2017 and July 2018 (wet season), the Chao Phraya River Mouth received high freshwater inflows, $21.4\text{-}62.9 \times 10^6$ m^3/d, but the contamination of heavy metals entering the river mouth was diluted – the load per unity of volume, 0.657-0.659 μg/L. Consequently, the input of total heavy metals to the river mouth was high, 14.059-41.254 kg/d. In January 2019 (dry season), the Chao Phraya River Mouth received low freshwater inflows, 8.3×10^6 m^3/d, and the contamination of heavy metals entering the river mouth was diluted – the load per unity of volume, 0.667 μg/L. Consequently, the input of total heavy metals to the river mouth was very low, 5.537 kg/d. The variation of total heavy metals to the Chao Phraya River Mouth was observed, most probably related to seasonal river flows and heavy metal contamination levels in water.

Table 3 Inputs of total heavy metals to the Chao Phraya River Mouth during 2017 and 2019

Period of time	Inflow (m^3/d)	Input	Hg	Cd	Zn	Ni	Pb	TML
January 2017	9.1×10^6	µg/L	0.007	0.074	1.047	0.541	4.428	6.097
		kg/d	0.064	0.673	9.528	4.923	40.295	55.483
June 2017	62.6×10^6	µg/L	0.006	0.193	0.218	0.052	0.190	0.659
		kg/d	0.376	12.082	13.647	3.255	11.894	41.254
July 2018	21.4×10^6	µg/L	0.003	0.056	0.177	0.161	0.260	0.657
		kg/d	0.064	1.198	3.788	3.445	5.564	14.059
January 2019	8.3×10^6	µg/L	0.001	0.029	0.102	0.320	0.215	0.667
		kg/d	0.008	0.241	0.847	2.656	1.785	5.537

TML=total metal load

Conclusion

The enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in suspended sediment and bottom sediment of the Chao Phraya River Mouth during 2017 and 2019. The observed order of heavy metal concentrations in sediments of the Chao Phraya River Mouth was as follows: Pb > Ni > Zn > Cd > Hg in suspended sediment and Zn > Pb > Ni > Hg > Cd in bottom sediment.

The mean concentrations of all dissolved and total heavy metals (Hg, Cd, Zn, Ni and Pb) in water monitored at the Chao Phraya River Mouth during 2017 and 2019 were lower than the coastal water quality standard in Thailand. The observed order of dissolved heavy metal concentrations in water of the Chao Phraya River Mouth was as follows: Zn ≥ Cd ≥ Pb ≥ Ni > Hg.

The high levels of heavy metal concentrations in suspended and bottom sediments of the Chao Phraya River Mouth might be caused by weathering of rock through water and wind, suspended sediment transport to the river mouth by river flow, tide and seasonal monsoon, and bottom sediment deposition, especially at the inside of river bends and river mouths. Because of the high freshwater inflow in the Chao Phraya River and the discharge of low amount of heavy metals in the treated industrial effluent into the Chao Phraya River, possible sources of heavy metals in suspended and bottom sediments of the Chao Phraya River Mouth were not anthropogenic, but rather lithogenic.

The seasonal variation of heavy metals in suspended and bottom sediments of the Chao Phraya River Mouth was observed, related to the fast river flow in wet season, result in more suspended sediment transported to the river mouth or bottom sediment flushing. The spatial variation of heavy metals in suspended and bottom sediments of the Chao Phraya River was existed, due to adsorption and co-precipitation of dissolved heavy metals onto suspended and bottom sediments and aggregation-sedimentation of suspended sediment during transport.

The seasonal variation of dissolved heavy metals (in µg/L) in water of the Chao Phraya River Mouth was observed, due to accumulation and release of dissolved heavy metals from the river suspended and bottom sediments. The seasonal variation of total heavy metals (in kg/d) to the Chao Phraya River Mouth was observed, related to the river flows and heavy metal contamination levels in water.

Acknowledgement

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