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Contamination of Cadmium and Mercury along the East Coast of the Gulf of Thailand: A Comparative Study Using the Green Mussel (*Perna viridis*), Artificial Mussel, Water and Sediment

Varaporn Cholumpai^a, Praparsiri Kachanopas-Barnette^b*, SukandaTubmeka^c, Rudolf Shiu-Sun Wu^d

^aRegional Medical Sciences Center 6 Chonburi, Department of Medical Sciences, Ministry of Public Health, Thailand

^{b,c}Environmental Science Programme, Faculty of Science, Burapha University, Chonburi, Thailand
 ^{b,c}Center of Excellence on Environmental Health and Toxicology, Bangkok, Thailand
 ^bDepartment of Aquatic Science, Burapha University, Chonburi, Thailand
 ^dDepartment of Science & Environmental Studies, Education University of Hong Kong
 ^aEmail: varaporn.c@dmsc.mail.go.th, ^bEmail: praparsi@buu.ac.th

^cEmail: adnakus@hotmail.com, ^dEmail: rudolfwu@eduhk.hk

Abstract

We compared Cd and Hg concentrations in green mussel (*Perna viridis*) in wet and dry seasons along the Gulf of Thailand affected by different anthropogenic activities (Angsila - aquaculture, Sriracha - shipping, and Maptaphut - heavy industry). Levels of Cd in green mussels from Angsila during wet (mean \pm SD 1.72 \pm 0.20 μ g/g dry wt.) and dry seasons (1.21 \pm 0.52 μ g/g dry wt.) were significantly higher than those in Sriracha and Maptaphut. Likewise, levels of Cd in artificial mussel (AM) were also highest in Angsila. Mean concentration of Hg in green mussels fluctuated only within a narrow range (0.07- 0.08 μ g/g dry wt.) at all sites in the wet season, but was significantly lower in the dry season (0.03 μ g/g dry wt.). Maptaphut had the highest Hg concentration in sediment during the dry season (0.17 \pm 0.11 μ g/g dry wt.). In contrast, level of Hg in mussel was 2-3 times higher in the wet season than the dry season at all sites.

^{*} Corresponding author.

Nevertheless, levels of Cd and Hg in mussel and sediment at all sites were still below the national Seafood/Sediment quality standards established in other countries. No significant correlation was found between level of Cd in mussels with those in AM and sediment or between sediment and AM. This research has successfully used the AM for monitoring of Cd, indicating both mussels and AM can provide a reliable estimate on Cd contamination in the aquatic environment.

Keywords: heavy metal contamination; mussels; artificial mussels; seafood safety.

1. Introduction

Coastal areas of the Gulf of Thailand are at risk of heavy metal contamination from anthropogenic discharges from industries, agriculture, urban wastewater as well as shipping activities [1]. During the wet season, surface runoff from the watershed add further load of contaminants to coastal water. Notably, the eastern of Thailand (Chonburi and Rayong) are popular tourist areas. Since the mid 1980s, the economies of these two provinces have transformed from agriculture-based to industrial-based, which in turn has caused rapid deterioration of coastal water quality and heavy metal contamination. Evidently, level of cadmium and mercury in seawater, sediment, and bivalves has shown an increase in the coastal waters around Chonburi and Rayong provinces. Cd was only measured at 0.01 - 0.26 μ g/L in seawater during 1981 – 1982 [2], 0.38 - 1.53 μ g/g dry wt. in green mussel (P. viridis) from 1974 – 1995 [3], and 0.07 - 0.72 mg/kg wet wt. in blood cockle (Anadara granosa) along the Chonburi coast from 2015 – 2016 [4]. Thongra-ar and Parkpian (2002) found concentrations of Hg in coastal areas of Thailand were generally within safe level from 1974 - 1999, except for a few samples that exceeded the standard [1]. Similarly, a mercury concentration in blood cockle from the Chonburi coast was 0.0036 - 0.0120 mg/kg wet wt. in 2015 - 2016 [4] Overall, concentrations of cadmium and mercury in seawater, sediment and marine organisms in the Gulf of Thailand have been found to fall within the standards and guidelines of Thailand as well as other countries. The green mussel is the most important species cultured along the coast of Thailand, contributing to around 44 percent of total production of coastal aquaculture by weight, which is the highest production in Thailand [5]. Green mussel (Perna viridis) is widely cultured along the east coast of Thailand for human consumption. The fact that cadmium and mercury can be biomagnified along the food chain may pose a public health risk to the consumers. The remarkable ability of green mussels to concentrate heavy metals from diet and water makes them a good choice for bio-monitoring of heavy metals [6]. Traditionally, heavy metal concentration in the aquatic environment can be estimated by chemical analysis of water, sediments, and aquatic organisms. However, each method presents its own limitations and difficulties. For example, metal in water is typically low with large temporal variations, making sampling and analysis difficult and not cost effective. Metals in sediment typically confounded by sediment characteristics (e.g. particle size, organic content, and redox condition) which cannot be unified and compared between sites. Metal accumulation in aquatic organisms is often species specific, and also significantly affected by a great variety of physical factors (e.g. salinity, temperature, tidal level, pollution level) and biological factors (e.g. growth and reproductive stages, food availability) [7, 8]. These confounding factors make it difficult to compare metal levels in biomonitors under different hydrographic conditions and seasons. Moreover, the limited natural distributions of species often prevent comparison over large geographic areas. To overcome the above limitations of biomonitors, Wu and his colleagues (2007) have developed a chemical device known as the

artificial mussel (AM) [9]. Earlier laboratory and field studies have shown that AM can accumulate Cd, Cr, Cu, Pb, Zn and Hg from water in a concentration-dependent manner, including the bioavailable fraction, and therefore can provide a time-integrated estimate of metal concentrations in the environment. The AM can be employed in contrasting environmental conditions including highly polluted water and therefore confers a significant advantage in field monitoring. Subsequently, AM has been successfully used in field monitoring and evaluation of heavy metal contamination in marine and freshwater environments including Hong Kong, Scotland, Iceland, Portugal, Australia, South Africa, China, Korea and Bangladesh [1, 9, 10, 11, 12, 14, 15]. This study compared Hg and Cd in green mussels, AM, sediment and water at three green mussel culture sites along the coast of Chonburi and Rayong provinces (Gulf of Thailand) with different anthropogenic activities (i.e. Angsila, a well-known aquaculture site; Sriracha, a major shipping port and Maptaphut, one of the largest industrial areas in Thailand). The aim is to determine and compare contamination levels of Cd and Hg amongst these three sites in marine environments.

2. Materials & Methods

2.1 Study sites

Three sites were selected from two provinces along the eastern coast of Gulf of Thailand for the current investigation (Figure 1). Maptaphut, a site in Rayong Province, is characterized by heavy industrial establishments that depend on natural gas for manufacturing and production, while local power plants rely on coal to generate electricity. The important industries are gas separation, petrochemicals, chemical fertilizer, oil refining, steel, and products made from steel. The second site is Sriracha in Chonburi Province, which is highly urbanized, and where the largest seaport of Thailand is situated. The seaport is used for loading and unloading cargo from bulk carrier boats for international shipping. Examples of goods being transported include coal dust, cassava chip, cement, sulfur powder, fertilizers, soda ash, and gypsum powder. In contrast, Angsila is famous for mollusk culture due to its location in the Bangpakong River. However, coast waters of Angsila is directly influenced by the agricultural activities (cattle farming, shrimp culture, fishery, poultry and pig farms, and rice fields) in the watershed of the Bangpakong River, with an area of 18,500 km². Commercial culture of the green mussel (P. viridis) is common at all the above-three sites. In this study, mussel samples were collected from areas near mussel farms at each site. Two sampling areas of each site were selected. The Maptaphut site covers 3 km (coordinates are M1 N 1401298 E 735564; and M2 N 1401552 E 734976). The Sriracha site covers 5 km (S1 1457717 E 708141; and S2 N 1456295 E 707234). And finally, Angsila site covers 5 km (A1 N 1476463 E 708051; and A2 N 1474429 E 706194) as shown in Figure 1.

2.2 Sample collection and preparation

Samples of green mussels, sediment, seawater and AMs were collected during the dry season (April - May) and wet season (October - November) of 2016. Green mussels (umbo lengths of 6 - 7 cm) were collected from each site and transferred directly to the laboratory and maintained in aquaria with clean sand-filtered sea water before further metal analysis. Soft tissue from 20 mussels from each site were pooled into one sample (where n = 10) and freeze dried at - 40 °C. 0.5 ± 0.01 g freeze dried tissue was then extracted with a mixture of super pure

NHO₃:H₂O₂, 5:2 v/v and digested by Block Digestion System Model AIM600 at 110 °C for 6 hours. The extract was diluted with deionized water and adjusted to a volume of 50 ml and analyzed for Cd and Hg. Ten surface sediment samples were collected from each site and season using a peterson grab and transported to the laboratory in a cooler. Each sediment sample was sieved, grounded, freeze dried at - 40 °C and digested. Samples for Cd analysis were digested using a mixture of aqua regia (HNO₃ and HCl, 1:3 ratio) [16], while samples for Hg analysis was digested using a mixture of HNO₃:HCl, 9:1 (v/v) [17]. At each site, one liter of seawater was collected from 1 meter below the surface with a non-metallic water sampler (Model 1080 series GO-FLO). pH of each seawater sample (10 replicates of water sample collected from the field) was adjusted to 3 by adding 240 µl of nitric acid before analysis (by in-house method TE-CH-038, based on Standard Methods for Examination of Water and Wastewater) [18]. The samples were used for analyzing Cd concentrations.



Figure 1: Map of the Gulf of Thailand, showing the three study sites and the six sampling locations (Angsila and Sriracha, Chonburi Province, and Maptaphut, Rayong Province).

Artificial mussels (AM) were prepared in the laboratory following the method of Wu and his colleagues (2007) [9]. Each AM consisted of a Perspex tube (60 mm \times 25 mm), containing 0.2 g Chelex-100[®] resin, BioRad suspended in artificial seawater and sealed with polyacrylamide gel at both ends. Ten AMs were put inside a nylon bag housed by a plastic cage (200 mm \times 270 mm) and deployed 1 meter below water surface from the raft of a mussel farm. After 30 days, the AMs were retrieved and the Chelex resin in each device was emptied into a sintered glass filter and rinsed several times with Milli-Q water. Following the method of Wu and his colleagues (2007) [9], the resin was eluted in a 6 M nitric acid solution, which was prepared with nitric solution and 70% supra-pure. The elutriant from the Chelex resin was then diluted with Milli-Q water and made up to a known volume, and used for analyzing concentrations of Cd and Hg. Green mussels and sediment samples were

analyzed for Cd and Hg concentrations, while only Cd was analyzed in seawater and AM. Dissolved oxygen (DO), water temperature, conductivity, salinity and pH were also measured at each site during the field deployment and sampling using a multi-parameter water quality devices (brand YSI Pro2030 and pH meter D-71 from Horiba LAQUA).

2.3 Cadmium and mercury analysis

The precision and accuracy of heavy metal measurements in this study were assessed using standard reference material samples. For green mussel, we used SRM 2976, freeze-dried mussel tissue from the National Institute of Standards and Technology, MD, USA. The obtained results were in good agreement with the certified values of Cd and Hg at 0.820 ± 0.160 and 0.061 ± 0.036 mg/kg dry wt., respectively. Recovered values were determined by range of % recovery, where Cd was between 102 - 105% and Hg was between 103 - 111%. For marine sediment, we used MESS-3 from the National Research Council of Canada (Certified Values of Cd and Hg were 0.240 ± 0.010 and 0.091 ± 0.009 mg/kg dry wt., respectively). The % recovery for Cd was between 82 - 93% and Hg was between 106 - 115%. The AM device was only used for measuring the amount of Cd. Quality control was performed by comparing Cd amount to a standard curve created by Cd of different concentrations, which resulted in a linear regression function with a confidence interval $\mathbb{R}^2 \ge 0.995$. The quality control of seawater was tested using the standard spiked sample method, which resulted in the % recovery of Cd between 95 - 110% and Hg between 92 - 96%. Cd from green mussel, sediment and the AM were determined using Graphite Furnace-Atomic Absorption Spectrometer (Varian, model SpectrAA-640Z). Cd in seawater samples were analyzed using Inductively Coupled Plasma Mass Spectrophotometer (ICP-MS), model 7500c. Hg in green mussel and sediment were analyzed using ICP-MS (Agilent, model 7500c).

2.4 Statistical analysis

The results were expressed as means \pm SD. Values were normally distributed after transformation. The Anderson-Darling test for normality was conducted on the data prior to performing the two-way ANOVA (2 factor analysis of variance in CRD) followed by the Tukey's test. For all statistical tests, differences were considered significant at p < 0.05. The statistical software Minitab17 was used and graphs were plotted with Microsoft EXCEL 2007. Correlation of cadmium levels in green mussel, sediment and the AM was analyzed using Spearman Rho's two-tailed analysis (coefficient, r), while mercury level was compared between green mussel and sediment.

3. Results

3.1 Cadmium concentrations in green mussel, sediment, seawater and the AM device

Concentrations of Cd in mussels, sediment, seawater and AM at the three study sites are shown in Table 1 and Figure 2. Cd concentration in mussel tissue at Angsila during the wet season $(1.72 \pm 0.20 \ \mu\text{g/g} \text{ dry wt.})$ was significantly higher than those from other sites (p < 0.05). Cd concentrations in sediment were also significantly higher at Angsila during the wet season ($0.17 \pm 0.03 \ \mu\text{g/g} \text{ dry wt.}$) and Maptaphut during the dry season ($0.20 \pm 0.13 \ \mu\text{g/g} \text{ dry wt.}$) compared with other sites. AM from Angsila contained the highest concentration of Cd (0.03

 \pm 0.01 µg/g of Chelex) during the dry season which was significantly higher than other sites (p < 0.05). However, Cd was below detection limit (< 0.6 µg/L) in all the sixty samples of seawater (n = 60) from all three sites in both dry and wet seasons. No correlation was found between levels of Cd in mussel and sediment (r = -0.124, p = 0.344); between mussel and AM (r = -0.015, p = 0.911); or between sediment and AM (r = -0.011, p = 0.932).

 Table 1: Cadmium concentrations (mean ± SD) in green mussel, sediment, and artificial mussel from the three cities in eastern Thailand during dry and wet seasons in 2016.

Season	Green mussel (µg/g dry wt.)		Sediment (µg/g dry wt.)		AM ($\mu g/g$ of Chelex)	
	Mean \pm SD	Range	Mean \pm SD	Range	$Mean \pm SD$	Range
Dry	1.21±0.52	0.67-1.89	0.07 ± 0.01	0.05-0.09	0.03 ± 0.01	0.02-0.04
Wet	1.72±0.20	1.31-1.93	0.17±0.03	0.12-0.22	0.01 ± 0.01	ND*-0.03
Dry	0.63±0.09	0.49-0.75	0.07 ± 0.02	0.04-0.12	0.01 ± 0.01	ND*-0.02
Wet	0.34±0.12	0.19-0.61	0.11 ± 0.02	0.08-0.14	0.01 ± 0.01	ND*-0.03
Dry	0.05 ± 0.01	0.04-0.06	0.20±0.12	0.04-0.36	0.01 ± 0.01	ND*-0.02
Wet	0.09 ± 0.01	0.06-0.11	0.14 ± 0.10	0.05-0.28	0.02 ± 0.00	0.01-0.02
	Season Dry Wet Dry Wet Dry Wet	Green musse Mean ± SD Dry 1.21±0.52 Wet 1.72±0.20 Dry 0.63±0.09 Wet 0.34±0.12 Dry 0.05±0.01 Wet 0.09±0.01	$\begin{array}{r c} Season & \hline Green mussel (\mu g/g dry wt.) \\ \hline Mean \pm SD & Range \\ \hline Dry & 1.21 \pm 0.52 & 0.67 - 1.89 \\ \hline Wet & 1.72 \pm 0.20 & 1.31 - 1.93 \\ \hline Dry & 0.63 \pm 0.09 & 0.49 - 0.75 \\ \hline Wet & 0.34 \pm 0.12 & 0.19 - 0.61 \\ \hline Dry & 0.05 \pm 0.01 & 0.04 - 0.06 \\ \hline Wet & 0.09 \pm 0.01 & 0.06 - 0.11 \\ \hline \end{array}$	$\begin{array}{c c} Season & \hline Green mussel (\mu g/g \ dry \ wt.) & Sediment (\mu g/g \ dry \ wt.) \\ \hline Mean \pm SD & Range & Mean \pm SD \\ \hline Dry & 1.21 \pm 0.52 & 0.67 - 1.89 & 0.07 \pm 0.01 \\ \hline Wet & 1.72 \pm 0.20 & 1.31 - 1.93 & 0.17 \pm 0.03 \\ \hline Dry & 0.63 \pm 0.09 & 0.49 - 0.75 & 0.07 \pm 0.02 \\ \hline Wet & 0.34 \pm 0.12 & 0.19 - 0.61 & 0.11 \pm 0.02 \\ \hline Dry & 0.05 \pm 0.01 & 0.04 - 0.06 & 0.20 \pm 0.12 \\ \hline Wet & 0.09 \pm 0.01 & 0.06 - 0.11 & 0.14 \pm 0.10 \\ \hline \end{array}$	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $

ND* : $< 0.0125 \ \mu g/g$ of Chelex



Figure 2: Concentrations of cadmium in green mussel tissue (μ g/g dry wt.), sediment (μ g/g dry wt.) and the artificial mussel (μ g/g of Chelex) from three sites in eastern Thailand in dry and wet seasons (2016). Significant differences (p < 0.05) among sites and seasons are indicated by different letters, with a > b > c > d > e > f.

3.2 Mercury in green mussel and sediment

In green mussel tissue, the highest Hg concentration was found at Maptaphut during the wet season ($0.08 \pm 0.01 \mu g/g dry wt$.), which is significantly higher than that in the dry season (p < 0.05). Likewise, Hg concentrations

at the other two sites during the wet season were significantly higher than those in the dry season, (p < 0.05), as shown in Table 2 and Figure 3. On the other hand, the sediment samples from Maptaphut in the dry season (0.17 \pm 0.10 µg/g dry wt.) contained the highest levels of Hg, and represented the only statistically significant difference (p < 0.05) among the samples, as shown in Table 2 and Figure 3. Moreover, positive correlation was found between the Hg concentrations in green mussel and sediment, with coefficient r = -0.354, p = 0.005.

Table 2: Mercury concentrations (mean \pm SD) in green mussel and sediment from three sites in easternThailand during dry and wet seasons (2016).

Site Location	Season	Green mussel (µ	g/g dry wt.)	Sediment (µg/g dry wt.)	
		Mean ± SD	Range	Mean ± SD	Range
Angsila	Dry	0.03±0.01	0.02-0.05	0.03±0.01	0.02-0.06
	Wet	0.07±0.02	0.04-0.10	0.02±0.02	0.01-0.06
Sriracha	Dry	0.03±0.00	0.03-0.03	0.03±0.01	0.02-0.05
	Wet	0.08±0.02	0.06-0.09	0.01±0.00	0.01-0.02
Maptaphut	Dry	0.03±0.00	0.03-0.04	0.17±0.10	0.03-0.30
	Wet	0.08±0.01	0.07-0.09	0.03±0.02	0.01-0.07



Figure 3: Concentrations of mercury in green mussel samples (μ g/g dry wt.) and sediment (μ g/g dry wt.) from three sites in eastern Thailand during dry and wet seasons (2016). Significant differences (p < 0.05) among sites and seasons are indicated by different letters, with a > b > c.

Seawater parameters were measured at each site, and results are shown in Table 3. The data show conductivity in the wet season to be lower than in the dry season. During the dry season, Angsila had the highest conductivity (57.6 μ S/cm) and salinity levels were higher in the dry season (26.0 - 32.5 psu) than in the wet season (23.6 - 28.4 psu)

Season	Site	DO(mg/I)	Temperature (°C)	Conductivit	Salinity	ъЦ
Season	Site	DO (IIIg/L)	remperature (°C)	y (µS/cm)	(psu.)	pn
Dry Season	Angsila	4.9	31.1	57.6	32.0	7.3
	Sriracha	4.8	30.9	50.6	32.5	7.6
(March - April, 2016)	Maptaphut	2.9	32.6	46.4	26.0	7.9
Wet Season	Angsila	3.1	30.4	48.9	25.5	8.0
	Sriracha	3.8	31.4	42.9	28.4	6.3
(October - November, 2016)	Maptaphut	4.6	29.4	44.0	23.6	6.4

Table 3: In-situ water quality parameters at three sites in eastern Thailand, during dry and wet seasons (2016).

4. Discussion

4.1 Cadmium concentrations in green mussel, sediment, seawater and AM devices

Cd in green mussel from Angsila during the wet season $(1.72 \pm 0.2033 \ \mu g/g \ dry \ wt.)$ was 2 - 34 times higher than other sites, in both seasons. The level of Cd found in this study was higher than corresponding values reported nationwide and from other countries, as shown in Table 5. For example, the Cd value in green mussel at the Rayong River mouth ranged from 0.38 - 1.35 mg/kg dry wt. (1974 - 1995), Cd in green mussel from Angsila was 1.53 mg/kg dry wt. (1995) [3], or 3 times higher than green mussel tissue from Hong Kong at 0.66 μ g/g dry wt. [19] and Muar Estuary, Johore, Malaysia (2006) at 0.58 μ g/g dry wt. [20]. However, the Cd concentrations in mussels found in this study were lower than permissible safety limits set by Australia [21] and Brazil [22]. Standards/guidelines for Cd in seafood in Thailand have yet to be developed. Cd contamination levels in sediment at Maptaphut during the dry season ($0.20 \pm 0.12 \mu g/g dry wt$.) and at Angsila during the wet season (0.17 \pm 0.03 µg/g dry wt.) were 2-3 times higher than other sites. In addition, this result is also 2 - 3 times higher than Cd levels found in sediment 12 years ago at the same site [23]. The levels of Cd in sediment from the three sites along the east coast of Thailand in this study and from past research were lower than the Sediment Quality Standard of Thailand, which should be less than 2 mg/kg dry wt. [24], and they were lower than the levels found in sediment from the Hong Kong coast [25] as well as the Guidelines of Australia and New Zealand [26], as shown in Table 5. However, this comparison is a bit tricky since metal level in sediment would depend on particle size and % organic content. Mussels have been used extensively as a sentinel biomonitor for heavy metals in the aquatic environment [27,28,29,30]. In the present study, Cd accumulation was found in both green mussel and the AM. Despite both mussels and AM detected high levels of Cd in Angsila during the dry season, overall, no significant correlation could be found between Cd levels in green mussels and the AM devices among sampling sites and seasons. The lack of correlation between Cd in green mussel (P. viridis) and AM is similar to research reported from the coastline of China, where Cd levels were not correlated, while other metals such as Hg, Pb, Zn, Cu, and Cr were significantly correlated. The reason may be that AM can only take up dissolved phase metals, while live mussels can take up both dissolved phase of metal from water and also particulate metals from food [14]. This research is the first to use the AM in Thailand, and Cd levels (ranged from < 0.01 - 0.04 ug/g of chelex) are comparable with those found in AM studies in South Africa [31], Australia [12], and China [14]. However, the Cd levels from this research are 10 - 75 times lower than Cd levels

found in Hong Kong [9], Scotland and Iceland [10], Portugal [11], Korea [13], and Bangladesh [15] as shown in Table 5. Notably, Cd levels found in seawater were $< 0.6 \,\mu$ g/L at the three sites in the Gulf of Thailand, which is lower than Cd level found in seawater reported in the same area from $1981 - 1982 (0.01 - 0.26 \mu g/L)$ [2]. Nevertheless, Cd in water is lower than the standards set by the European Commission [32] as well as Guidelines of Australia and New Zealand [26], as shown in Table 5. The results from this study show the highest Cd level from Angsila for both wet and dry seasons from sediment, green mussel, and the AM device. Angsila is characterized by an open bay connected to the Bangpakong River, which received discharges from a watershed covering 18,500 km², of which 60% is agricultural land [33]. Farmers in the watershed may use chemical fertilizers, herbicides, fungicides, and phosphate fertilizers, which contain cadmium as one of the key ingredients [34]. Moreover, sludge generated from urban and industrial waste water is often used to produce organic fertilizer by the agriculture industry, and may form another source of cadmium contamination [35]. Cd contamination problems around the Angsila area must be regularly monitored and evaluated, agricultural, animal farm (shrimp, fish, pig and poultry) and other anthropogenic discharges from the watershed to the Bangpakong River must be tightly controlled or else Angsila area may not be suitable for aquaculture culture in the future. Although Maptaphut is an industrial area, Cd level contamination was found to be low in the present study, which agrees with the results of regular wastewater quality monitoring conducted by the Department of Industrial Works. Finally, Cd contamination was not found in the Sriracha seaport because shipping activities do not form a significant source of Cd contamination.

4.2 Mercury concentration in green mussel and sediment

For all three sites (Maptaphut, Angsila, and Sriracha) in the wet season, Hg concentrations in green mussel were 2 - 3 times higher than in the dry season. In 2016 (the present study period), the rainfall was heavier than usual (1,805.3 mm for the wet season and 107.7 mm for the dry season) [36]. This could have caused more Hg to be carried in watershed surface runoff, as well as mixing of sediment into the water column from strong winds, leading to increased uptake of Hg by the mussels. The correlation between green mussel and sediment confirms the negative correlation and indicates that lower levels of Hg can be found in sediment, but higher levels of Hg can be found in green mussel due to its accumulation in green mussel tissue. In contrast, Hg concentrations in green mussel from this study were lower than that revealed in the green mussel (P. viridis) from the Bangpakong River mouth (0.095 µg/g dry wt.) in 1998 and the Rayong River mouth (0.170 µg/g dry wt.) [37]. The Hg concentrations in green mussel (P. viridis) in this study were also lower than the Standard of the US Food and Drug Administration (1.25 mg/kg dry wt.) [38]. As for Hg concentrations in sediment, the Hg concentration from Maptaphut during the dry season ($0.1715 \pm 0.1048 \ \mu g/g \ dry \ wt$) was 5 - 11 times higher than the other two sites, and represented a three-fold increase from 1998 - 2004. However, the value is still lower than the Sediment Quality standards of Thailand, of 0.4 mg/kg dry wt. [24] and lower than the Sediment Hg Guidelines of Florida (0.13 - 0.7 mg/kg dry wt.) [39], Hong Kong (0.5 - 1.0 mg/kg dry wt.) [40], and Australia and New Zealand (0.15 - 1.0 mg/kg dry wt.) [26] as shown in Table 6. As expected, Hg contamination in green mussel and sediment from Maptaphut was higher than that in Angsila and Sriracha, because Maptaphut is an industrial area that produces chemical products, chemical fertilizer, iron and steel, and contains oil refineries, a coal power plant, and a center for elimination of hazardous materials. From previous monitoring, the concentration of mercury in sediment (35 cm depth) was in the range of 0.15 - 1.61 mg/kg dry weight [41],

which is higher than the standards of Thailand and other countries. Since Maptaphut was included in the Pollution Control Announcement in 2009, Hg contamination in sediment and green mussel around Maptaphut is no longer a problem. The research from this study found levels of Hg in sediment and green mussel from all three sites lower than standards of Thailand and other countries.

5. Conclusions

The result of this study showed Cd contamination in green mussel tissue, sediment, and the AM device around Angsila. The high concentration found in green mussel tissue, suggest that regular monitoring during the rainy season, as well as measures for reducing anthropogenic discharges into the Bangpakong River, are deemed necessary. This research has successfully used the AM for monitoring and found high level of Cd contamination in the water column: a similar result was found in the green mussel tissue analysis. The highest concentration of Hg was found in green mussel during the wet season, and the highest level of sediment Hg was found in Maptaphut during the dry season.

6. Recommendations

A continuous monitoring and evaluation of Cd contamination in aquatic life around Angsila is compulsory, because this area has a higher possibility to find Cd in the marine ecosystem. Since green mussel (*P. viridis*) is a commercially species commonly consumed by the local population at a large amount the high level of Cd and Hg found in mussels call for an urgent health risk assessment.

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 Table 5: Comparison of Cd concentrations from mussel (P. viridis), sediment, seawater and the artificial mussel (AM) device in this study and studies from other Asian countries. Standards and Guidelines from selected countries are also presented.

Years of study	Location	Mussel (ug/g dry wt.)	Sediment (ug/g dry wt.)	Sea water (ug/L)	The AM (ug/g of chelex)	Reference
2016	Angsila, Chonburi	0.67-1.93	0.05-0.22	ND	ND*-0.04	Present study
2016	Sriracha, Chonburi	0.19-0.75	0.04-0.14	ND	ND*-0.03	Present study
2016	Maptaphut, Rayong	0.04-0.11	0.04-0.36	ND	ND*-0.02	Present study
1974- 1995	Rayong	0.38-1.35	-	-	-	Ruangwises and Ruangwises (1998) ³
1981– 1982	The Upper and the Lower Gulf of Thailand	-	-	0.01-0.26	-	Hungspreug (1982) ²
1995	Chonburi	1.53	-	-	-	Ruangwises and Ruangwises (1998) ³

1998	Laem Chabang	-	< 0.1	-	-	PCD (1999) ⁴²
1998	Map Ta Phut	-	< 0.1-0.2	-	-	PCD (1999) ⁴²
2004	Industrial Estate Bangpakong River - Ang Sila,	-	0.024-0.19	-	-	Thongra-ar and his colleagues (2008) ²³
2004	Chon Buri Laem Chabang, Chon Buri	-	0.008-0.15	-	-	Thongra-ar and his colleagues $(2008)^{23}$
2004	Map Ta Phut, Rayong	-	< 0.006- 0.17	-	-	Thongra-ar and his colleagues $(2008)^{23}$
1998- 2004	Hong Kong Coast	-	0.1-5.3	-	-	Zhou and his colleagues $(2007)^{25}$
No data	Victoria Harbor, Hong Kong	-	-	-	0.4-3.0	Wu and his colleagues $(2007)^9$
1998- 1999	Scotland and Iceland	-	-	-	0.1-3.5	Leung and his colleagues (2008) ¹⁰
2006	Muar Estuary, Johore, Malaysia	0.58	-	-	-	Kamaruzzaman and his colleagues $(2008)^{20}$
2007- 2008	Portugal	-	-	-	> 0.05-1.0	Rey and his colleagues $(2011)^{11}$
2009	South Africa	-	-	-	0.00-0.031	Degger and his colleagues $(2011)^{31}$
2009– 2011	Victoria, Australia	-	-	-	0.01-0.013	Kibria (2012) ¹²
No data	Korea	-	-	-	2.5-3.5	Ra and his colleagues $(2014)^{13}$
No data	China	-	-	-	0.033-0.618	Degger and his colleagues $(2016)^{14}$
No data	South Africa	-	-	0.001-0.002 mg/L Fresh water	0.001-0.0016 mg/kg of	Claassens and his colleagues (2016) ⁴³
2013 Standard	Bangladesh	-	-	-	0.3-0.5	Kibria (2016) ¹⁵
Australian	Legal	10.0 (mg/kg	-	-	-	NHMRC (1987) ²¹
Brazilian Ministry of Health		5.00 (mg/kg drv wt.)	-	-	-	ABIA (1991) ²²
European Commission		-	-	2.5	-	$EC(2001)^{32}$
Australia and New Zealand		-	1.5-10 (mg/kg dry wt)	< 0.5-5	-	ANZECC and ARMCANZ (2000) ²⁶
Thai Coas Quality St	tal Sediment andard	-	2.0 (mg/kg dry wt.)	-	-	PCD (2015) ²⁴

 $ND = non detectable level at < 0.6 \mu g/L$

 ND^{\ast} = non detectable level at $< 0.0125~\mu\text{g/g}$ of Chelex

Years of	Location	Mussel	Sediment	Reference
study		(ug/g dry wt.)	(ug/g dry wt.)	
2016	Angsila, Chonburi	0.02-0.10	0.01-0.06	Present study
2016	Sriracha, Chonburi	0.03-0.09	0.01-0.05	Present study
2016	Maptaphut, Rayong	0.03-0.09	0.01-0.30	Present study
1998	Bangpakong River	0.095	-	Chongprasith and
	Mouth			Wilairatanadilok (1999) ³⁷
1998	Rayoung River Mouth	0.170	-	Chongprasith and Wilairatanadilok (1999) ³⁷
1998	Laem Chabang Industrial Estate	-	< 0.005-0.032	PCD (1999) ⁴²
1998	Map Ta Phut Industrial Estate	-	< 0.005-0.172	PCD (1999) ⁴²
2004	Bangpakong River- Ang Sila, Chon Buri	-	0.018-0.121	Thongra-ar and his colleagues $(2008)^{23}$
2004	Laem Chabang, Chon Buri	-	0.010-0.117	Thongra-ar and his colleagues $(2008)^{23}$
2004	Map Ta Phut, Rayong	-	0.007-0.116	Thongra-ar and his colloagues $(2008)^{23}$
1998-2004	Hong Kong Coast	-	0.05-8.0	Zhou and his colleagues (2007) ²⁵
Standard/Gu	ideline			
Thai Coastal S Standard	Sediment Quality	-	0.4 (m g/kg dry wt.)	PCD (2015) ²⁴
Standard of th Drug Adminis	e USA Food and	1.25 (mg/kg dry wt.)	-	USFDA (1990) ³⁸
Australia and	New Zealand	-	0.15-1.0 (mg/kg dry wt.)	ANZECC and ARMCANZ (2000) ²⁶ ; Food Standards Australia (2001) ⁴⁴ ; NRS (2012) ⁴⁵
Hong Kong		-	0.5-1.0 (mg/kg dry wt)	HKGS (1998) ⁴⁰
Florida		-	0.13-0.7 (mg/kg dry wt.)	MacDonald (1994) ³⁹

Table 6: Comparison of Hg concentrations from mussel (*P.viridis*), sediment and sea water in this study, studies from other Asian countries and Standard Guideline.

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