Arsenic pollution assessment in surface sediment of the inner Gulf of Thailand

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Abstract. Surface sediments of the inner Gulf of Thailand and four main river estuaries were collected for total arsenic analysis in order to assess spatial distribution pattern, contamination status, regulating factors and potential ecological risk. The study reveals that the arsenic concentrations in the surface sediment samples varied from 5.43–18.01 mg/kg dry weight. Higher arsenic concentrations were found in the Chao Phraya river estuary toward in the eastern of the inner Gulf of Thailand. Based on the sediment quality guidelines (SQGs), arsenic was investigated most unlikely to cause any adverse effects. The result of geo-accumulation index (Igeo) was found to be less than 1, which indicated "unpolluted" state. Moreover, the potential risk of individual metal (Er) values directed that arsenic at all stations posed low ecological risks.

1. Introduction
Arsenic toxicity has become a principal concern owing to the escalating contamination of air, soil, water, and food. Millions of people in developed and developing countries are being chronically exposed to elevated doses of arsenic from natural and anthropogenic sources. Arsenic has the ability to readily change its oxidation state and bonding configuration, thus showing diverse chemical behavior in the environment and forming large numbers of organic and inorganic compounds. Natural source of arsenic is in the earth’s crust as present in geothermal discharges. Arsenic sources from human activities include mining [1] burning of coal, use of arsenic compounds in production processes, use of arsenical fungicides, insecticides, and herbicides in agriculture [2]. Arsenic occurs mainly in four oxidation states or inorganic arsenic, arsenate (As⁵⁺), arsenite (As³⁺), arsenic (As⁰) and arsine (As⁻³) [3]. After accumulation in biosphere media, the arsenic contamination is discharged by runoff into the marine environments. Subsequently, sediments act as a major sink for arsenic in the marine environments. On the contrary, sediment can be a potential source of arsenic and causes the process of biotransformation and bioaccumulation in marine ecosystems.

Marine sediments comprise significant sinks of contaminants and represent potential sources of pollution to the marine environment. Sediments from estuaries and coasts receiving drainage from industries and metal-mining areas contain significantly elevated arsenic concentrations. These contaminants are the threats of the aquatic biota and have been of significant environmental concern [4,5]. The studies of arsenic pollution in sediment have increased in recent years [6-9]. According to arsenic contamination, low arsenic concentrations in water and sediments could rise to hazardous levels through food chains in aquatic organisms by effects of bioaccumulation and biomagnification [10-11].
The inner Gulf of Thailand is due mainly to a variety of sources including surface runoff (particularly major large rivers including Mae Klong, Tha Chin, Chao Phraya and Bang Pakong Rivers) and drainage from port areas, domestic and industrial effluent discharges through outfalls and various contaminants from ships. The presence of arsenic contamination can potentially affect the inner Gulf of Thailand and ultimately becomes a serious environmental issue in Thailand.

The objectives of this study were (1) to investigate heterogeneity distributions of arsenic contamination in surface sediment of the inner Gulf of Thailand, (2) to determine the relationship between regulating factors and behavior of the arsenic, and (3) to measure the status and assess ecological risk of arsenic contaminations on the inner Gulf of Thailand.

2. Materials and methods

2.1. Study area and sampling

The inner Gulf of Thailand is the coastal environment, where is the transitional area between land and sea, and located at latitude 13°20'N, longitude 100°45'E. Its total shoreline is approximately 350 km long. Concerning the width of the inner Gulf of Thailand, the east to west and north to south is around, which covering an approximate area of 90×90 km² [12] with 15 m of average depth. The inner Gulf of Thailand receives large amounts of pollutions from point sources and non–point sources. In particular, runoff from Mae Klong (MK), Tha Chin (TC), Chao Phraya (CY) and Bang Pakong (BK) River estuaries in the northern part. Surface sediment (0-1 cm) samples were collected in July 2017, from 59 locations of the inner Gulf of Thailand and estuaries including Mae Klong, Tha Chin, Chao Phraya and Bang Pakong River estuaries (Figure 1) using the Ekman or Smith-Mcintyre stainless steel grab sampler. The surface sediments were sliced off and quickly placed in acid washed 50 ml of polyethylene centrifuge tube. All samples were preserved in the dark container filled with ice (below 4 ºC). Laboratory analyses were then carried out as quickly as possible.

2.2. Chemical analysis

All apparatus were previously soaked in 20% (v/v) HNO₃ solution for 24 h and were rinsed with double-distilled water before used. Total surface sediment analyses the decomposition of the sediment sample by strong acids. Homogenized sediment samples (0.5 g) were digested in acid-cleaned Teflon vessels containing 4 ml of concentrated HF, which were allowed to react overnight at room temperature. Subsequently, 2 ml of aqua regia (HCl:HNO₃ at a ratio 3:1 v/v) were added to the sample. Samples were digested for 15 min at 200 ºC using high performance microwave digester (Milestone Series 135931, Italy). After cooling, 2.0 g of H₃BO₃ were added to dissolve the fluoride precipitates. After that, the supernatant was poured in to a PP volumetric flask and were diluted to 50 ml with De-ionized reverse osmosis water [13,14]. All digested samples were analyzed total arsenic using hydride
generation technique of Atomic Absorption Spectrophotometer (AAS: Agilent 240AA, U.S.A.). The relative accuracy for the determination of arsenic was analyzed within the National Research Council Canada (NRCC) sediment reference material MESS-4. For data validation, the accuracy and precision of the arsenic analytical method was being acceptable at 74.0%. For readily oxidizable organic matter was determined using chromic acid method [15]. The total organic carbon (TOC) and total nitrogen (TN) were determined in part of the samples after removal of carbonate with 2 N HCl (60 ºC, 24 h) using a CHN Elemental Analyzer (JM 10, J-SCIENCE LAB Co., Ltd Kyoto JAPAN). For acid volatile sulfide was analyzed using 18 N H2SO4 and measured by Gastec model 201 LH.

2.3. Data processing

Sediment quality guidelines (SQGs)

The SQGs by Canadian and Wisconsin were popularity in the world because as a result of three sets were directly applied (without normalization) in assessing possible arsenic contamination in sediment of the study area. With respect to Canadian and Wisconsin SQGs, a site is determined as contaminated the reliability of the threshold effect level (TEL) and probable effect level (PEL) [16].

Geo-accumulation Index ($I_{geo}$)

The index of geo-accumulation was used to separate the anthropogenic effects on the sediment from the natural influences [17]. The $I_{geo}$ is defined by the following equation:

$$I_{geo} = \log_2 \left( \frac{\text{Sample}}{1.5 \times \text{Background}} \right)$$

where sample is the concentration measured in the sediment and background is the preindustrial level as reported by Hakanson [18]. The $I_{geo}$ has 7 classifications following: <0 is practically unpolluted, 0–1 is unpolluted to moderately polluted, 1–2 is moderately polluted, 2–3 is moderately to strongly polluted, 3–4 is strongly polluted, 4–5 is strongly to very strongly polluted, and >5 is very strongly polluted [19,20].

Ecological risk assessment

In this study, the evaluation of ecological risk for a given metal was made by adopting the potential risk of individual metal ($E_r$) [20].

$$E_r = T_{r1} \times C_i$$

Where $E_r$ is potential risk of arsenic, $T_{r1}$ is 10 of toxic-response factor for arsenic [18]. Classifications for $E_r$ as follow: $E_r < 40$ is low potential ecological risk, $40 \leq E_r < 80$ is moderate potential ecological risk, $80 \leq E_r < 160$ is considerable potential ecological risk, $160 \leq E_r < 320$ is high potential ecological risk, and $E_r \geq 320$ is very high ecological risk.

$$C_i = \frac{c_i}{c_{r1}}$$

where $C_i$ is contamination factor, $c_{r1}$ is mean concentration of the metal for study and $c_{r1}$ is 15 of preindustrial reference values in the sediment [21].

3. Results and discussion

3.1. Spatial distribution

The spatial distribution of arsenic entire the inner Gulf of Thailand is shown in Figure 2. The arsenic concentrations ranged from 5.43-18.01 mg/kg dry weight. The maximum arsenic concentration was observed in the sediment samples of the Chao Phraya River estuary. On the contrary, the Mae Klong River estuary was the site with the lowest arsenic concentration. The result from all stations, the horizontal distributions of arsenic in the surface sediment was characterized by higher concentrations, which were found in the east of inner Gulf of Thailand. According to distribution pattern of arsenic is decreasing trend toward the lower area of the western. The high concentrations zone of arsenic are shown the Chao Phraya River estuary and extending eastward. Moreover, high concentrations of arsenic increased in the middle of the Gulf. In general, mean±SD concentrations of arsenic in the surface sediment were ranked, in decreasing order as follows: CY (11.25±3.88 mg/kg) > BK (8.66±1.00 mg/kg) > TC (8.02±1.28 mg/kg) > the inner GoT (7.46±2.23 mg/kg) > MK (7.13±1.65...
mg/kg) (Figure 2). Arsenic concentrations in this study (5.43-18.01 mg/kg) were higher than Africa (0.2-2.44 mg/kg) in some aquatic environments, but lower than Asia (0.42-342 mg/kg), Australia (21.2 mg/kg), Europe (21.2 mg/kg) and South America (1-27 mg/kg) in other locations (Table 1).

![Figure 2. The map shows the spatial pattern of arsenic concentration across the inner Gulf of Thailand and four major river estuaries generated by ordinary kriging method. The box plot compares the arsenic concentrations for all sampling locations with published data. The black line represents Coastal sediment quality standards of Thailand (SQS), arsenic standard level of 7 mg/kg.](image)

### Figure 2.

#### Table 1. Comparison of total arsenic content in sediment between this study and other studies mentioned in the literature.

| Locations                          | Descriptive statistical parameters | Total arsenic concentrations (mg/kg) | References |
|------------------------------------|------------------------------------|-------------------------------------|------------|
| Open Lagoon, Southwest Nigeria     | Mean                               | 2.44                                | [22]       |
| Dar es Salaam coast, Tanzania      | Range                              | 0.2–1.3                             | [23]       |
| Chabahar Bay, Iran                 | Range (mean±SD)                    | 5–22 (12.2±5.23)                    | [24]       |
| Southern part of Caspian Sea, Iran | Range                              | 7.17–13.52 (9.94±1.71)              | [25]       |
| East Kalimantan, Indonesia         | Mean                               | 2.00                                | [26]       |
| Arabian Gulf, Saudi Arabia         | Range                              | 53–342                              | [27]       |
| Bohai Sea, China                   | Range                              | 3.4–13.6                            | [28]       |
| Gorgan Bay South Caspian Sea, Iran | Range                              | 2.6–8.6                             | [29]       |
| West Bengal, eastern part, India   | Range (mean±SD)                    | 4.41–11.46 (5.85±1.20)              | [30]       |
| Southern Caspian Sea, Iran         | Range (mean±SD)                    | 7–17 (10.37±2.71)                   | [9]        |
| Peninsular, Malaysia               | Range                              | 21.81–59.49                         | [19]       |
| Southern/ Northern Gulf of Aqaba, Saudi Arabia | Mean                   | 15.1/12.2                           | [31]       |
| Eastern Beibu Bay, China           | Range (mean±SD)                    | 2.40–23.09 (9.53±3.99)              | [32]       |
| Thale Noi/                         | Range (mean±SD)                    | 5.7–10.8 (8.2±1.7)/                 | [33]       |
| Inner - Middle Lake/ Outer Lake,  | Range (mean±SD)                    | 3.7–10.8 (5.9±1.5)/                 | [33]       |
| Songkhla Lake System, Thailand     | Range                              | 5.1–25.7 (10.7±5.5)                 |            |
| Caspian Sea, Iran                  | Range (mean±SD)                    | 6.97–20.1 (12.5±3.04)               | [34]       |
| Caspian Sea, Azerbaijan            | Range (mean±SD)                    | 8.87–22.6 (14.7±4.15)               | [34]       |
| Caspian Sea, Russia                | Range (mean±SD)                    | 0.42–6.71 (2.97±1.95)               | [34]       |
| Deception Bay, Queensland, Australia | Mean±SD                       | 21.2±1.1                           | [7]        |
| Algeciras Bay, Spain               | Range (mean±SD)                    | 8–23 (11±5)                         | [35]       |
| Todos os Santos Bay, BA, Brazil    | Range                              | 1–27                                | [6]        |
| **The inner Gulf of Thailand**     | Range                              | **5.43-18.01**                      | **This study** |

### 3.2. Arsenic contamination status
The SQGs values for arsenic classification of the samples based on these guidelines are shown (Table 2). The results of classifying sediments based on the TEL and PEL values suggested that arsenic in surface sediment of all sites would rarely (<TEL, TEL-PEL and >PEL values) be expected to non-cause effects on biota. Based on the SQGs by Canadian, arsenic has a mean concentration from sampling sites in Mae Klong River estuary shows result <TEL (62.50%) and TEL-PEL (37.50%). For Tha Chin (77.78%) and Bang Pakong River estuaries (88.89%) would be classified as presenting %TEL-PEL more than <TEL, while Chao Phraya River estuary had 100% TEL-PEL. The inner Gulf of Thailand shows result <TEL (70.83%) and TEL-PEL (29.17%). In comparison to the Wisconsin SQGs, a mean arsenic concentration in Mae Klong River estuary, 87.50% was lowered than TEL, in which 12.50% of samples fall in the range between TEL and PEL. For 88.89% were lowered than TEL, 11.11% of Tha Chin and Bang Pakong River estuaries samples fall in the range between TEL and PEL. The samples of Chao Phraya River estuary shows result 55.56% were lowered than TEL, in which 44.44% of samples fall in TEL-PEL. Arsenic concentration in the inner Gulf of Thailand has 83.33% <TEL and 16.67% fall in TEL-PEL (Table 2). Thus, at concentrations of arsenic in surface sediments greater than the TEL, toxic effects from long term exposure to arsenic would be predicted to occur. While, the Igeo method was used to calculate the metal contamination levels in the surface sediment of the inner Gulf of Thailand. The Igeo values were ranged from -2.05 to -0.32, which were indicated that the surface sediments of the inner Gulf of Thailand are categorized as practically unpolluted level (Figure 3).

Table 2. Comparison between arsenic concentrations and numerical Sediment Quality Guidelines (SQGs) in mg/kg

|                  | MK River estuary | TC River estuary | CP River estuary | BK River estuary | The inner GoT |
|------------------|-----------------|-----------------|-----------------|-----------------|--------------|
| Min              | 5.43            | 6.11            | 7.53            | 7.19            | 5.76         |
| Max              | 10.18           | 10.80           | 18.01           | 10.28           | 14.48        |
| Average          | 7.13            | 8.02            | 11.25           | 8.66            | 7.46         |
| SD               | 1.65            | 1.28            | 3.88            | 1.00            | 2.23         |
| Canadian         | TEL=7.24        | PEL=41.6        |                 |                 |              |
|                  | %<TEL           | 62.5            | 22.22           | 11.11           | 70.83        |
|                  | %TEL-PEL        | 37.5            | 77.78           | 100             | 29.17        |
|                  | %>PEL           | 12.5            | 11.11           | 44.44           | 16.67        |
| Wisconsin        | TEL=9.80        | PEL=33.0        |                 |                 |              |
|                  | %<TEL           | 87.5            | 88.89           | 55.56           | 88.89        |
|                  | %TEL-PEL        | 12.5            | 11.11           | 44.44           | 16.67        |
|                  | %>PEL           | 12.5            | 11.11           | 44.44           | 16.67        |

3.3. Regulating factors of As contamination

Generally, sediment characteristics play an important role in interpreting the distribution of arsenic in marine sediments such as grain size, inorganic and organic pollutants [29,34]. The present study, OM, TOC, TN and AVS are additionally analyzed in order to determine the regulating factors of arsenic contamination in the inner Gulf of Thailand. The results showed that OM, TOC, TN and AVS contents varied in the ranges of 0.16-3.77%, 2.03-36.5 mg C/g, 0.30-4.56 mg N/g and 0.00-1.86 mg S/g, respectively (Table 3). Subsequently, Pearson correlations are performed in all data of arsenic, OM, TOC, TN and AVS (Table 3). Arsenic content did not significant related with sediment characteristics. As a result, non-significant correlations indicated that arsenic contaminations in surface sediment of the inner Gulf of Thailand are independent from OM, TOC, TN and AVS content. However, an excellent correlation exists between TOC and TN in the surface sediment of this system. A general correlation between TOC and TN has been established by many authors [36-38]. Correlation analysis gives a very high coefficient of determination ($r$=0.955, $p$<0.01), which suggested that the concentration of TN may be regulated by organic sources. Moreover, positive significant correlation based on the content of OM ($r$=0.497, $p$<0.01) and TN ($r$=0.503, $p$<0.01) with AVS was found in the surface sediment of the inner Gulf of Thailand, which was indicated that the progressive enhancement of the processes of anaerobic decomposition, with increasing organic matter and total nitrogen.
Table 3. Product moment of Pearson’s correlation between arsenic concentration and related parameters in surface sediment of the inner Gulf of Thailand.

| Parameters | As (mg/kg) | OM (%) | TOC (mg C/g) | TN (mg N/g) | AVS (mg S/g) |
|-----------|------------|--------|--------------|-------------|--------------|
| Range     | 5.43-18.0  | 0.16-3.77 | 2.03-36.5   | 0.30-4.56   | 0.00-1.86    |
| Mean±SD   | 8.26±5.27  | 1.99±0.95 | 12.9±6.99   | 1.70±0.97   | 0.27±0.48    |
| As (mg/kg) | 1.000      | ns      | ns           | ns          | ns           |
| OM (%)    | ns         | 1.000   | 0.559**     | ns          | 0.503**      |
| TOC (mg C/g) | ns      | 0.559** | 1.000        | ns          | ns           |
| TN (mg N/g) | ns       | 0.675** | 0.955**     | 1.000        | ns           |
| AVS (mg S/g) | ns       | 0.497** | ns          | ns          | 1.000        |

** is significant at the 0.01 level (2-tailed), ns is non-significant.

3.4. Ecological risk assessment
The single $E_r$ has been used to assess the potential ecological risk in coastal ecosystems [19,39]. In comparison of the present study, lower $E_r$ values were observed in the Mae Klong River estuary (3.62-6.78) and the inner Gulf of Thailand (3.84-9.66). While, higher $E_r$ values were observed in Chao Phraya River (5.02-12.01) and Tha Chin River (4.07-7.02) estuaries. However, the overall $E_r$ values were in the range of 3.62–12.01 (Figure 3), which were lower than 40 and indicated that, arsenic at all stations posed low ecological risks.

Figure 3. Comparison of the $I_{geo}$ and the $E_r$ of arsenic in the surface sediment of the inner Gulf of Thailand and four major river estuaries.

4. Conclusion
The present study provides a comprehensive baseline data of arsenic in the surface sediments collected from 59 sampling sites in the inner Gulf of Thailand and four important estuaries. Based on the SQGs, arsenic investigated were most unlikely to cause any adverse effects. The surface sediment from all sampling sites was not polluted by the studied values. The $E_r$ values based on the classifications by Hakanson, arsenic in the inner Gulf of Thailand on the surface sediment was shown as low ecological risk. The result of this study suggested low ecological risk, yet, 66.10% of all sampling locations were found with exceeded national standard limit. Thus, the arsenic treatment or monitoring devices should be provided in specific locations to reduce and mitigate ecological and health risks.

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