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## Arsenic Cadmium and Lead Concentrations in Sediment and Biota from Songkhla Lake: A Review

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### Abstract

This article reviews the existing data on trace elements (Arsenic (As), Cadmium (Cd) and Lead (Pb)) in sediments and biota from Songkhla Lake, a shallow coastal lagoon in Southern of Thailand between 1998 and 2012. At present, the situation of those trace elements in sediments show moderate concentration levels for Cadmium and Lead. High levels for Arsenic have been found in the Outer Section of Songkhla Lake especially at the mouths of Samrong, U-Taphao and Phawong canals. The accumulation of trace elements in fish muscle tissue was well within limits for human consumption.

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### 1. Introduction

Songkhla Lake (Fig. 1), a shallow coastal lagoon, is located in the Southern part of Thailand and has a total area of 1,042 km<sup>2</sup>. The lake itself is a very important natural resource for people who live in Songkhla Province, Phatthalung Province and some parts of Nakhon Si Thammarat Province as a major producer and nursery ground for local fisheries. The lake is divided into 4 sections; Thale Noi (27 km<sup>2</sup>), Inner Lake (473 km<sup>2</sup>), Middle Lake (360 km<sup>2</sup>) and Outer Lake (182 km<sup>2</sup>). The salinity ranges from fresh to saline. The outer section is open to the sea. It is deeper in the Inner Section (more than 2 m), quite shallow in the Middle section (around 1 m) and

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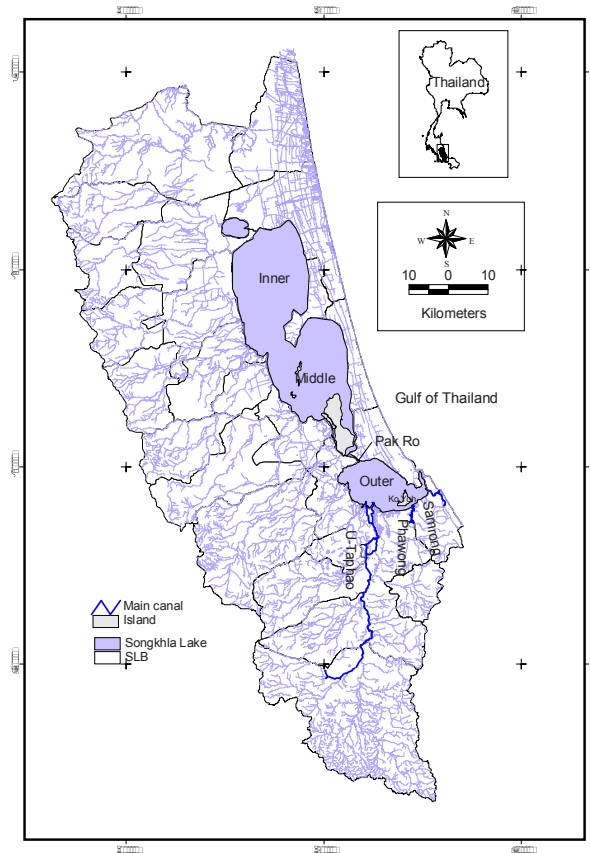


Fig.1 The map of Songkhla Lake.

Arsenic, Cadmium and Lead are recognized as toxic pollutants. Arsenic (As) is used in paints, dyes, metals, drugs, soaps, semiconductors and also is used as wood preservative. High arsenic levels can also come from certain fertilizers and animal feeding operations. Industrial activities such as copper smelting, mining and coal burning also contribute to arsenic in our environment. Cadmium (Cd) is used in the steel industry, plastic,

batteries, which is released to the environment via wastewater, fertilizers and local air pollution. The main sources of cadmium in the air are burning of fossil fuels such as coal or oil and the incineration of municipal waste (WHO, 2004) [1]. While lead (Pb) is exposed to human occurs primarily through drinking water, airborne and lead-based paints. The route of input trace metals to the marine environment are mainly via air and water. Atmospheric inputs are particularly important for the input into the open ocean. In the case of entry via water, it must be noted that most of the contaminants that enter the coastal lagoon or enter the seawater are initially present in fresh water.

During the past two decades, quite a number of studies concerning the area have been carried out but quite limited number of studies have been made regarding the pollution of the lake by heavy metals (As, Cd and Pb) in sediments and biota since 1996. The aim of the article is to review an existing data on the trace elements concentrations in sediments and biota in Songkhla Lake. The data are taken from various studies since 1996 up to present in order to document the past and present status of As, Cd and Pb in Songkhla Lake.

### Trace elements in Sediment

In the aquatic environment trace elements are distributed between the dissolved phase, colloids, suspended matter and sedimentary phases. Sediments and soils have a high storage capacity for contaminants. In the hydrological cycle, less than 0.1% of the metals are actually dissolved in the water and more than 99.9% are stored in sediments and soils (Salomons, 1998 [2]; Pradit et al., 2010 [3]). There have been not too many studies on As, Cd and Pb concentrations in sediment from Songkhla Lake during the past fifteen years. Maneepong and Rakaew (1998) [4] took the bottom sediment from different parts of the lake to analyze for the heavy metals. The results showed that the Pb concentrations were 13.0-46.3 mg kg<sup>-1</sup> dry wt and the Cd concentration were <0.1-0.7 mg kg<sup>-1</sup> dry wt. Meesuk (1999) [5] evaluated the concentrations of As, Pb and Cd in bottom sediment of Songkhla Lake. The findings revealed that Pb, Cd and As ranged between 26.55- 92.75, 0-1.25 and 0.2-2.50 mgkg<sup>-1</sup> dry wt, respectively. Maneepong and Angsupanich (1999) [6] took seven bottom sediment samples from the outer section of Songkhla Lake. All samples were analyzed for As and Pb. The result showed that levels of As and Pb ranged between <0.5-17.9 mgkg<sup>-1</sup> dry wt and between 0.5 - 28.2 mgkg<sup>-1</sup> dry wt, respectively. A high concentration of As was found in U Taphao canal. However Bhongsuwan and Bhongsuwan (2002) [7] reported that the concentrations of Cd in sediments from 4 sites in the Outer Songkhla lake was non-detectable but the concentration of Pb in the sediment samples were in ranges of 24.5-59.8 mgkg<sup>-1</sup> dry wt. After that, Sirinawin and Sompongchaikul (2005) [8] reported that total Pb concentrations in core sediment samples along the U-Taphao canal were 16.7 - 43.1 mgkg<sup>-1</sup> dry wt vary to a certain extent vertically and seaward. However, Nakinchart (2006) [9] studied the distribution of some heavy metals in surface sediments from Songkhla Lake and found that Cd levels were <1.3 mgkg<sup>-1</sup> dry wt in the whole part of the Lake while Pb levels were 3.5±0.6 mgkg<sup>-1</sup> dry wt in the Inner Lake; 4.0±2.2 mgkg<sup>-1</sup> dry wt in the Middle Lake and 3.8±1.3 mgkg<sup>-1</sup> dry wt in the Outer Lake. Sompongchaiyakul and Sirinawin (2007) [10] reported that As concentrations in sediment from Thale Noi, Inner & Middle Lake, and Outer Lake were 8.2 ± 1.7 (5.7-10.8), 5.9 ± 1.5 (3.7-10.8) and 10.7 ± 5.5 (5.1-25.7) mg kg<sup>-1</sup>, respectively. Pradit et al. (2010) [3] took the bottom sediments from 44 stations in Songkhla Lake to study the trace element contaminations in sediments. The trace element concentrations in surface sediment ranged from 0.8 - 70.7 µg g<sup>-1</sup> dry wt for As, 0.1 - 2.4 µg g<sup>-1</sup> dry wt for Cd and 8.2-131 µg g<sup>-1</sup> dry wt for Pb. The result showed that the Outer Section of the lake, in particular, the sediments at the mouths of Phawong, Samrong and U-Taphao canals were significantly enriched in trace elements due to municipal, agricultural and industrial discharges entering the lake through the canals. Samrong canal is an important source of Cd and Pb, whereas contamination of As is principally transported by Phawong and U-Taphao canals. Pradit et al. (2012) [11] reported that the higher concentrations of Cd and Pb in the surface sediments (0–8 cm) from the Outer section of the lake were found in Samrong canal. The concentrations in sediment ranged from 31-35 µg g<sup>-1</sup> dry wt for Pb, 0.21 – 0.28 µg g<sup>-1</sup> dry wt for Cd and 20.0-22.0 µg g<sup>-1</sup> dry wt for As. This is in agreement with previous results (Pradit et al. 2010) [3] which showed that Samrong canal is severely polluted with these trace metals. Higher concentrations of As is found in Phawong Canal. U-Taphao Canal is also enriched in As, but not in the other trace elements.

According to the US EPA toxicity classification, these sediments can be classified as moderately polluted for most of the studied metals, but heavily polluted for As.

Considering the data obtained from those reports beginning in 1998, it can be observed that higher Cd, As and Pb concentrations in sediment were increasing especially in the outer section of Songkhla Lake due to urban expansion and industrialization. The comparison of trace element concentrations (As, Cd and Pb) in sediments from Songkhla Lake was shown in Table 1.

Table 1 The comparison of trace elements concentrations (As, Cd and Pb) in sediments, and biota from Songkhla Lake

|   | As        | Cd          | Pb          | References                            |
|---|-----------|-------------|-------------|---------------------------------------|
| <b>Sediment (mg/kg dry wt.)</b>                 |           |             |             |                                       |
| Songkhla Lake                                   | -         | <0.1-0.7    | 13.0-46.3   | Maneepong and Rakaew (1998)           |
| Songkhla Lake                                   | 0.2-2.50  | 0-1.25      | 26.55-92.75 | Meesuk (1999)                         |
| Songkhla Lake: Outer Section                    | 1.7-32.6  | -           | -           | Maneepong and Angsupanich (1999)      |
| Songkhla Lake: Outer Section                    | -         | -           | 24.5-59.8   | Bhongsuwan and Bhongsuwan (2002)      |
| Songkhla Lake: Outer Section                    | -         | -           | 16.7-43.1   | Sirinawin and Sompongchaiyakul (2005) |
| Songkhla Lake: Inner Section                    | -         | -           | 3.50        | Nakinchat (2006)                      |
| Songkhla Lake: Middle Section                   | -         | <1.3        | 4.00        | Nakinchat (2006)                      |
| Songkhla Lake: Outer Section                    | -         | -           | 3.80        | Nakinchat (2006)                      |
| Songkhla Lake: Thale Noi                        | 5.7-10.8  | -           | -           | Sompongchaiyakul and Sirinawin (2007) |
| Songkhla Lake: Inner & Middle Section           | 3.7-10.8  | -           | -           | Sompongchaiyakul and Sirinawin (2007) |
| Songkhla Lake: Outer Section                    | 5.1-25.7  | -           | -           | Sompongchaiyakul and Sirinawin (2007) |
| Songkhla Lake                                   | 0.8 -70.7 | 0.1-2.4     | 8.2-131     | Pradit et al (2010)                   |
| Songkhla Lake                                   | 20-22.0   | 0.21-0.28   | 31-35       | Pradit et al (2012)                   |
| <b>Biota (µg/g dry wt.)</b>                     |           |             |             |                                       |
| <b>Fish</b>                                     |           |             |             |                                       |
| <i>Arius maculatus</i> : muscle tissue          | 0.50-7.24 | 0.00-0.03   | 0.06-0.40   | Pradit et al (2010)                   |
| <i>Arius maculatus</i> : liver                  | 0.27-1.96 | 0.09-1.37   | 0.24-3.44   | Pradit et al (2010)                   |
| <i>Arius maculatus</i> : fish egg               | 0.33-1.11 | 0.01-0.03   | 0.02-0.12   | Pradit et al (2010)                   |
| <i>Osteogeneiosus militaris</i> : muscle tissue | 0.19-0.60 | 0.00-0.01   | 0.05-0.18   | Pradit et al (2010)                   |
| <i>Osteogeneiosus militaris</i> : liver         | 0.26-0.62 | 0.10-0.53   | 0.14-0.87   | Pradit et al (2010)                   |
| <i>Oreochromis mossambicus</i> : muscle tissue  | 13.00     | -           | 3.00        | Maneepong and Angsupanich (1999)      |
| giant seaperch                                  | 0-0.250   | 0-0.010     | 0.163-1.985 | Meesuk and Benjakul (1998)            |
| <b>Shimp</b>                                    |           |             |             |                                       |
| <i>Macrobachium rosenbergii</i>                 | 2.00      | -           | 8.00        | Meesuk and Benjakul (1998)            |
| gaint tiger prawn                               | 0-0.150   | 0-0.380     | 0-2.625     | Meesuk and Benjakul (1998)            |
| <b>Mollusc</b>                                  |           |             |             |                                       |
| green mussel                                    | 0-1.750   | 0.040-0.343 | 1.548-1.985 | Meesuk and Benjakul (1998)            |
| <i>Corbicula arata</i>                          | 16.00     | -           | 10.00       | Maneepong and Angsupanich (1999)      |

## Metals enrichment factors

Regional background values assessed from core sediment samples are reported by Choi et al. (2008) [12] and offshore sediments in the Gulf of Thailand by Shazili et al. (1999) [13]. Background levels varied from 5-9  $\mu\text{g g}^{-1}$  dry wt for As, from 0.03-0.2  $\mu\text{g g}^{-1}$  dry wt for Cd, from 15-30  $\mu\text{g g}^{-1}$  dry wt for Pb. Averaged values normalized to Al were used to compare concentrations found at different stations to background values. Anthropogenic input of metals can be suspected when the normalized concentrations of metals in samples are higher than background levels (Valdés et al., 2005) [14]. Enrichment factors (EF) were calculated as  $\text{EF} = (\text{Me}/\text{Al})_{\text{sample}} / (\text{Me}/\text{Al})_{\text{background}}$ . The calculation of enrichment factors indicated that As and Cd concentrations in the outer section of Songkhla Lake are around 5-10 times higher than background concentrations (Pradit et al., 2010 [3]; Pradit et al., 2012 [11]). Especially concentration of As in the Outer section of the lake has been increasing since 1998. One of the sources of As may be the local wood industry, where As is used as a wood preservative (Sompongchaiyakul and Sirinawin, 2007) [10]. Major sources of waste discharged into the U-Taphao canal are from rubber, parawood, and seafood processing industries with a discharge rate of 41,000  $\text{m}^3/\text{day}$  (Sirinawin and Sompongchaiyakul, 2005) [8].

## Comparison with sediment quality guidelines (SQGs)

Several sediment quality guidelines (SQGs) using chemical and biological effect databases have been established. These SQGs are summarized in Table 2. NOAA presents ERL (effects range low) and ERM (effects range median) guidelines for estuarine and marine environments which represent the 10<sup>th</sup> and 50<sup>th</sup> percentiles of adverse biological effects (NOAA, 1999) [15]. The SQGs for freshwater environments (McDonald et al., 2000) [16] have a lower TEC (threshold effects concentration) and an upper PEC (probable effect concentration) at which toxicity to bottom dwelling organisms are predicted to be unlikely and probable, respectively. The Oslo and Paris Conventions for the prevention of marine pollution also established Ecological Assessment Criteria (EAC) (OSPAR, 1997) [17]. EAC values are the threshold values. A lower EAC value is derived from the protection of all marine species, also the most sensitive. A higher EAC value is the highest concentration that does not cause acute toxic effects. The USEPA has also made classifications (non-polluted, moderately polluted and heavily polluted) based on toxicity tests (Baudo et al., 1990 [18]; Filgueiras et al., 2004 [19]). Pradit et al. (2010) [3] reported that the Pb levels at the mouth of Samrong canal exceeded the PEC value and were classified as heavily polluted. As levels exceeding the PEC value were observed in the outer section of the lake. Cd levels exceeding the TEC value were observed only at the mouth of Samrong canal. In Samrong canal, the concentrations of As, Cd and Pb exceeded the ERL values. In U-Taphao and Phawong canals, the ERL value for As was exceeded. Comparing to the data from Pradit et al. (2012) [11], the As levels were classified as heavily polluted at the outer section of Songkhla Lake.

Table 2 Sediment quality guidelines (SQGs) based on effects to benthic-dwelling species ( $\text{mg kg}^{-1}$  dry wt)

|    | US EPA Toxicity classifications |                     |                  | OSPAR | MacDonald et al. |     | NOAA |     |
|----|---------------------------------|---------------------|------------------|-------|------------------|-----|------|-----|
|    | Non Polluted                    | Moderately Polluted | Heavily Polluted | EAC   | TEC              | PEC | ERL  | ERM |
| Pb | <40                             | 40-60               | >60              | 5-50  | 36               | 130 | 46.7 | 218 |
| As | <3                              | 3-8                 | >8               | 1-10  | 9.8              | 33  | 8.2  | 70  |
| Cd |                                 |                     | >6               | 0.1-1 | 0.99             | 5   | 1.2  | 9.6 |

Remarks EAC = Ecological Assessment Criteria, TEC = threshold effect concentration, PEC = probable effect concentration, ERL = effects range low, ERM = effects range median

### Trace elements in Biota

Aquatic organisms accumulate contaminants from the environment and therefore have extensively been used in pollution monitoring programs (UNEP, 1993) [20]. The accumulation pattern of contaminants in fish and other aquatic organisms depend both on uptake and elimination rates. Fish forms an important part of human food and thus numerous studies have been carried out on metal contaminations in different species of fish but quite a limited number of studies on contamination of heavy metals in fish have been done in Songkhla lake since 1998. Meesuk and Benjakul (1998) [21] studied amounts of arsenic and heavy metals in aquatic animals in Songkhla Lake. The findings revealed that there were As, Pb and Cd in giant seaperch ranging between 0-0.250, 0.163-1.985 and 0-0.010  $\mu\text{g g}^{-1}$  dry wt, respectively. There were As, Pb and Cd in giant tiger prawn between 0-0.150, 0-2.625 and 0-0.380  $\mu\text{g g}^{-1}$  dry wt, respectively. There were As, Pb and Cd in green mussel ranging between 0-1.750, 1.5548-1.985 and 0.041-0.343  $\mu\text{g g}^{-1}$  dry wt., respectively. The species seemed to play an important role in trace metals accumulation were bivalves. Maneepong and Angsupanich (1999) [6] reported that considerable variability of As concentrations was observed from lower than 2  $\text{mg kg}^{-1}$  dry wt in fish to 16  $\text{mg kg}^{-1}$  dry wt in molluscs. The highest value in fish-meat was found in *Oreochromis mossambicus* taken from Phawong canal (13  $\text{mg kg}^{-1}$  dry wt). The highest value in mollusk meat was found in *Corbicula arata* taken from U Taphao canal (16  $\text{mg kg}^{-1}$  dry wt). The concentrations of Pb in shrimp and molluscs tended to be higher than those found in fish but were considered similar to background. The concentrations of As, Cd, and Pb in muscle and liver as well as in the eggs of two catfish species (*A. maculatus* and *O. militaris*) caught in different sections of the lake was reported by Pradit et al. (2012) [11]. The concentrations of As, Cd, and Pb in muscle tissue of *A. maculatus* were in the ranges of 0.50–7.24, 0.00–0.03, and 0.06–0.40  $\mu\text{g g}^{-1}$  dry wt, respectively, whereas the concentrations in liver were in the ranges of 0.27–1.96, 0.09–1.37 and 0.24–3.44  $\mu\text{g g}^{-1}$  dry wt, respectively. In addition, The concentrations of As, Cd, and Pb in the fish eggs were 0.33–1.11, 0.01–0.03 and 0.02–0.12  $\mu\text{g g}^{-1}$  dry wt, respectively. The concentrations of As, Cd, and Pb in muscle tissue of *O. militaris* were in the ranges of 0.19–0.60, 0.00–0.01, and 0.05–0.18  $\mu\text{g g}^{-1}$  dry wt, while the concentrations in the liver were in the ranges of 0.26–0.62, 0.10–0.53 and 0.14–0.87  $\mu\text{g g}^{-1}$  dry wt. No significant difference in trace element concentrations between the two catfish species could be found. Cd essentially accumulates in the liver, whereas the distribution of Pb between liver and muscle tissue is highly variable. The concentrations of Cd, Pb and As found in fish eggs were lower than those found in muscle and liver tissue.

According to the European Union regulations, maximum allowable levels in fish are only provided for Hg, Cd and Pb (EC, 2001) [22], whereas in other countries, maximum allowable levels for As, Ni, Cr, Cu and Zn are also established (USEPA, 1989 [23]; FAO, 1983 [24]). The EU limit for Cd and Pb are 0.25  $\mu\text{g g}^{-1}$  dry wt and 2.5  $\mu\text{g g}^{-1}$  dry wt, respectively. Legal limits for As vary between 0.5 - 50  $\mu\text{g g}^{-1}$  dry wt. depending on whether the values are expressed as inorganic fraction or total As content. The Joint FAO/WHO Expert Committee (1983) set a limit of 0.5  $\mu\text{g g}^{-1}$  dry wt for inorganic As. In fish, trace elements may be concentrated at different levels in different organs of the body and may be present in different forms (inorganic and organic forms). High levels of arsenic are encountered in marine fish, but are predominantly present in the non-toxic arsenobetaine form (De Gieter et al., 2002 [25]; Baeyens et al., 2009 [26]). Inorganic As, on the other hand, is highly toxic and carcinogenic. From an existing data, levels of the elements observed in general are well within the maximum residual limits indicating that consumption of the fish from Songkhla Lake does not pose a threat to human health. At present, no work has been done on speciation of As in fish tissue to distinguish between toxic inorganic forms and nontoxic organic forms. Comparison of trace element concentrations (As, Cd and Pb) in Biota from Songkhla Lake was shown in Table 1.

### Conclusions

Studies of trace metal concentrations in sediments of Songkhla Lake showed moderate contamination levels of most metals except As which had high levels. Comparison with regional background values revealed a high enrichment factor for As and Cd in the outer section of the lake especially at the mouths of Samrong, U-Taphao

and Phawong canals. The anthropogenic inputs of trace metal for Arsenic was observed in the outer section of the lake. These are directly associated with point discharges from municipal, agricultural and industrial activities. Data from the diffusion fluxes which were calculated from the pore water profiles showed that there was a significant release of As from sediments, especially at the mouth of U-Taphao and Phawong canals (Pradit et al., 2012) [11]. However, the trace metal concentrations found in biota from Songkhla Lake were well within acceptable ranges for human consumption. The contaminations of As, Cd and Pb in biota should be concerned. A regular monitoring program especially on biota in the outer section of Songkhla Lake is recommended

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