Asian Journal of Water, Environment and Pollution, Vol. 4, No. 1, pp. 17-24.

Arsenic, Chromium and Mercury in Surface Sediment of Songkhla Lake System, Thailand

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Received October 10, 2005; revised and accepted November 15, 2006

Abstract: Songkhla Lake, a tropical estuarine lagoon system in southern Thailand, is located between latitudes 7°08′ and 7°50′ N and longitudes 100°07′ and 100°37′ E. The system has four interconnected water bodies, namely Thale Noi, Inner Lake, Middle Lake and Outer Lake, ranging from fresh to saline water. The system receives runoff, wastewater and sediments from surrounding watershed and drains into the Gulf of Thailand. Seventy-four surface sediment samples from the whole lake system were determined for As, Cr, Hg, Fe, Mn, Al, organic carbon and textural characteristics. Total concentration of As in Thale Noi, Inner & Middle Lake, and Outer Lake are 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^{-1}, respectively. Total concentration of Cr in Thale Noi, Inner & Middle Lake, and Outer Lake are 45.2 ± 5.0 (36.0-55.2), 36.6 ± 9.2 (23.7-73.3) and 29.6 ± 9.2 (7.9-48.2) mg kg^{-1}, respectively. Total concentration of Hg in Thale Noi, Inner & Middle Lake, and Outer Lake are 89.1 ± 14.9 (63.4-113.1), 38.4 ± 11.1 (24.3-68.0) and 49.3 ± 15.2 (26.6-110.3) µg kg^{-1}, respectively. In most part of the lake, As and Cr concentrations are at or near natural levels, except at station 57. The concentration of Hg may be natural or influenced to some degree by anthropogenic sources. Thale Noi, the most natural part of the lake system which contains high organic content, has the highest concentrations of Cr and Hg. Diagenetic processes involving iron oxides and aluminosilicate appear to be the most important factor controlling the behaviour of As, Cr and Hg in sediments. Organic matter and clay size particles seem to have less direct influence on the geochemistry of the metals.

Key words: Arsenic, chromium, mercury, sediment, factor analysis.

Introduction

Arsenic (As), chromium (Cr) and mercury (Hg) are among the metal pollutants that have become of evolving environmental concern lately. The presence of these ions in the environment is due to both natural weathering and human activities. These metals are transported to the marine coastal environment through rivers and the atmosphere. However, the relative influence of natural and anthropogenic sources on the geochemistry of marine sediments is not always clear.

Arsenic can exist in inorganic form, organic form and gaseous state. Inorganic species are highly toxic. In oxidised sediment arsenate is the dominant species present, and is associated primarily with iron oxyhydroxides. In reducing marine sediments arsenate is reduced to arsenite and is associated primarily with sulfide minerals (Neff, 1997). The behaviour of arsenic in natural waters is depending on pH and redox conditions; arsenic forms oxy-anions of the oxidation

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states +3 and +5. Biological processes may also produce methylated arsenic species (Shorin et al., 1997). The mobility and toxicity of arsenic thus so depend on redox condition and the availability of possible carrier phases. Fe(III)-oxyhydroxides are main carrier phases of sorbed arsenic (De Vitre et al., 1991; Kuhn & Sigg, 1993) and play an important role in the diagenetic cycle at the sediment-water interface (Belzile & Tessier, 1990; Widerlund & Ingri, 1995; Sullivan & Aller, 1996). Other particles, such as Mn-oxides, are also important carrier phases (Peterson & Carpenter, 1986).

Chromium in sediments may be presented in one of two thermodynamically stable oxidation states: Cr(III) or Cr(VI) (James & Bartlett, 1983). The Cr(III) has a potential to be oxidised to Cr(VI), which has high health risk, by Mn-oxides (Johnson & Xyla, 1991; Fendorf et al., 1992) or by molecular oxygen at pH >9 (Bartlett, 1998). The Cr(III), therefore, has a tendency to be adsorbed specifically on clay and oxide surfaces and to hydrolyse within common environmental pH ranges (pH 3-9) (Bartlett & James, 1988). Most Cr(III) in sediments therefore should occur as highly insoluble hydroxy polymers sorbed to colloid surfaces (Fendorf & Sparks, 1994). However, Dubbin (2004) found that organic ligands can be effective chelators of sorbed chromium. As such these ligands may play an important role in the mobilisation and cycling of chromium in sediments.

Mercury (Hg) is relatively ubiquitous in the environment and is found in almost every environmental compartment including air, volcanic gases, freshwater, seawater, soils, mineral ores, sediments, and living organisms. Sources of mercury are both natural, via weathering and degassing of the earth’s crust, and anthropogenic (D’Itri, 1990). Like other pollutants, mercury is present in several minerals that are mobilized and also by natural processes and finally transported to sediments. The sediments are often a major sink for mercury compounds in aquatic environments where it can be rendered virtually inactive (deep sediments) or converted to methyl-Hg, principally by sulphur-reducing bacteria (Bigham and Henry, 1993).

Study Site and Environmental Setting

Songkhla Lake is a tropical estuarine lagoon system, formed by an interaction of land and ocean processes over geological time, in southern Thailand (Figure 1). It is located between latitudes 7°08′ and 7°50′ N and longitudes 100°07′ and 100°37′ E. The system comprises four interconnected water bodies, namely Thale Noi,

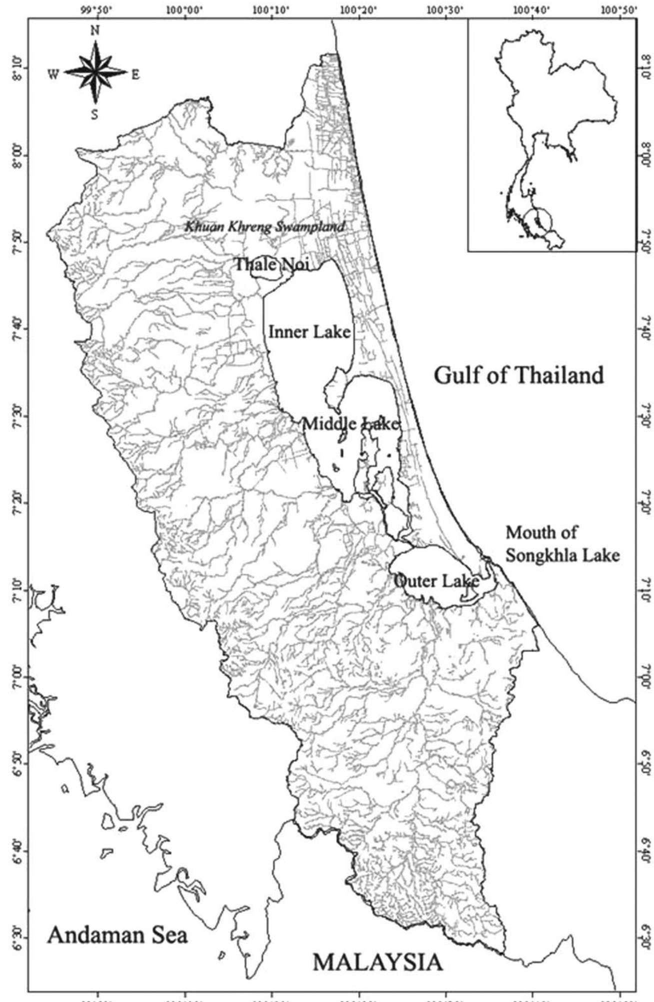

Figure 1: Locations map of Songkhla Lake system and river network.

Inner Lake, Middle Lake and Outer Lake, ranging from fresh to saline water. The lagoon system is connected to the Gulf of Thailand through a deep and narrow outlet at Songkhla city. The system receives runoff and wastewater from surrounding watershed. The runoff carries sediment onto the lakes, which will be transported through the lakes by the general movement of water towards the Gulf of Thailand. Sediment will also enter and leave the system via the outlet to the Gulf. According to EmSong Project (1998), the average residence time of watermass in the Inner, Middle and Outer Lakes are 55, 28 and 15 days, respectively. The average accumulation rate of sediment in the lake is 2.5-3.5 mm yr^{-1} which at some places is >15 mm yr^{-1} (PSU, 2004).

Thale Noi (27 km^2), located at the uppermost of the system, is a very shallow lake with an average depth of 1.2 m. It is a part of Khuan Khreng swampland (about

125 km^2). The Inner Lake is the biggest compartment with an average depth of 2 metres. It covers about 473 km^2. Tidal range in the Inner Lake is less than 0.09 m. The water is fresh all year round, with an exception of the dry year when salinity may increase up to 10 psu. The Middle Lake has an area of 360 km^2. The average depth is 1.5 m. This part of the Lake comprises several islands. Tidal range is about 0.11 m. Salinity is ranged from 0 to 20 psu depending on runoff (PSU, 2004). The Outer Lake has an average depth of 1.5 m with the exception of dredging channel which is about 12-14 m deep. The area is about 182 km^2. This part of the Lake system receives freshwater from the biggest and most urbanized and industrialized watershed of the basin. The salinity vary from 0 in wet season to 23-30 psu in dry season depending on proportion of freshwater runoff and inflow seawater. The tidal range at the outlet is about 0.6 m.

In recent years, considerable attention has been given to the environmental situation of Songkhla Lake system. Sources of pollutants to the lake system include unsanitary drainage from urban areas, industrial wastes (mainly related to rubber and food products), pollution from boat and fish wastes from Songkhla harbour, nearshore drainage, and municipal wastes from Hatyai and Songkhla cities. However, the status of trace elements in sediments of this lake system is not well known.

This paper presents the first spatial distribution of arsenic, chromium and mercury in sediments of the whole lake system. Total concentration of these metals, percent oxidizable organic carbon (%OC), as well as concentrations of iron (Fe), manganese (Mn) and aluminium (Al) were measured at a comprehensive network of sampling stations in order to gain an overview of the concentration, distribution and carrier of these metals in sediments.

Material and Methods

Seventy-four surface sediment samples (Figure 2) were collected using a Birke-Ekman Grab covering the whole lake system during October 2003 to January 2004. Station locations were determined by using the Global Positioning System (GPS) which is accurate to ±10 m. The samples were placed in zip-lock plastic bags and frozen immediately. In the laboratory, the samples were freeze dried and sub-sampled for sedimentological and chemical analyses.

The amount of sand (>63 μm), silt (2-63 μm) and clay (<2 μm) fractions were determined gravimetrically after wet sieving of the sample. Oxidizable organic carbon

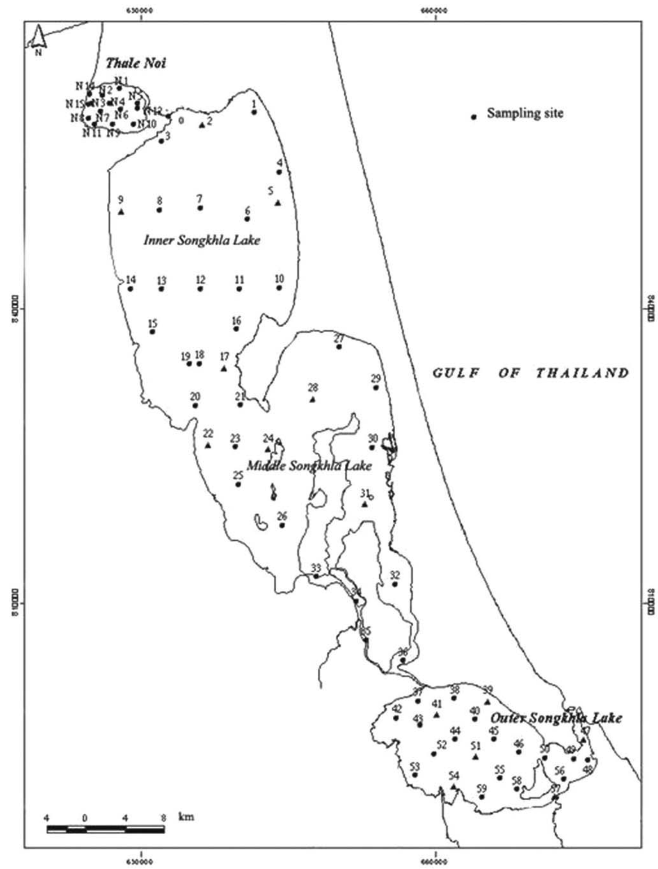

Figure 2: Surface sediment sampling stations in Songkhla Lake system.

were determined by modified Walkey-Black method (chromic method) as described in Loring and Rantala (1995).

For As determination, samples were prepared for analysis using the dry ashing method of Penrose et al. (1975). A portion of (~10-30 mg) of the dry powder was accurately weighed (± 0.01 mg) directly into a crucible and mixed with 1.5 ml of slurry freshly prepared from 30 g $Mg(NO_3)_2$, 50 g MgO and 500 ml of nanopure water. The mixture was dried overnight at 80°C and digested in a muffle furnace (200 °C for 1 hr, 300°C for 1 hr and 500 °C for 8 hr). The residue was dissolved in 6 M HCl (2.50 ml) and the solution mixed with 2.50 ml nanopure water. This solution is determined by HGAAS using Perkin-Elmer MHS-20 hydride system coupled with Perkin-Elmer 2380 AAS.

Other metals were determined by Perkin-Elmer Optima 2000 DV ICP-OES after microwave digestion. About 300 mg (± 0.01 mg) of the dried sediment (<63 μm) was digested with 6 ml HF and 1 ml of aqua regia in 40-ml LORRAN PTFE bombs (in a domestic type microwave oven), following the digestion technique described by Loring and Rantala (1995). Mercury concentration was determined using a Nippon Instruments Corporation Mercury Analyzer (Model SP-3D). A portion of dry sediment, accurately weighed to ± 0.01 mg, was decomposed by heating in the mercury atomizer. The Hg vapour was collected by two-step gold amalgamation process at 150°C. Elemental Hg was then liberated by heating to 700°C. The vapourised Hg was detected by the cold vapour atomic absorption technique.

Analytical precision of all sedimentological and chemical analyses was determined by analyzing every tenth sample in duplicate. The relative accuracy for metal analyses was within the standard deviation of the certified values for the National Research Council of Canada (NRCC) sediment reference material MESS-1 and BEST-1 (Hg only).

Principal component factor analysis of the correlation matrix was used to determine the variation of metal compositions, percent organic carbon and percent clay size particles in different sediment samples. The principal component analysis allows 'a factor score' for each sample to be calculated. When plotted by factor scores, samples with similar analyte compositions (scores) will be closer than those with dissimilar compositions. The results of the statistical valuation are used to suggest the geochemical processes controlling the accumulation and partitioning of the metals in the sediments.

Results and Discussion

Texture of Sediments and Organic Contents

The textural data indicates that 90% of the surface sediment in the lake system contained more than 95% of material size <63 μm. Oxidizable organic carbon in Thale Noi ranges from 2.9% to 9.2% with an average of 6.2 ± 1.8%, which is six times higher than those found in the main lake (Table 1). A spatial distribution of %OC accumulated in Songkhla Lake sediment is presented in Figure 3. It is clearly seen in that Khuan Khreng swampland might be a source of OC of the Inner Lake. In the Outer Lake, a slightly higher OC in the sediment at the inner part is probably due to the tidal effect.

Abundance of Trace Metals in Surface Sediments

All average concentrations and ranges of trace element in surface Songkhla Lake sediment are presented in Table 1.

Arsenic concentration in Songkhla Lake system is in the range of 4 to 26 mg kg^{-1}. Looking in details, high concentration of As was found at station 56, 57, 58 and 59 (Figure 2) at the range of 17 to 26 mg kg^{-1}. All the high value stations are located not far from the mouth of Pa-Wong canal which received industrial wastewater before emptying to Songkhla Lake. Station 57, which has the highest concentration, is located at the canal mouth. One of the industry that might be the source of As is the parawood preservation, using As and Cu compounds as wood preservatives, which is increasing in this area in the last decade. However, the concentration of As is still within the same range of uncontaminated estuarine sediments which contains from ~5-15 mg kg^{-1} dry weight total arsenic (Neff, 1997). Loring (1988) reported an average 6 mg kg^{-1} of As in sediments from the Gulf of St. Lawrence. Total As in sediment from Lake Macquarie, New South Wales, Australia, was found in the range of 2 to 20 mg kg^{-1} (Ellwood & Maher, 2003) and 4 to 17 mg kg^{-1} (Roach, 2005).

Table 1: Statistical data for textural characteristic, oxidizable organic carbon content, and concentration of arsenic, chromium, mercury, iron, manganese and aluminium in surface sediments of Songkhla Lake

	Sand (%)	*Silt (%)*	*Clay (%)*	*OC (%)*	*As (mg kg^{-1})*	*Cr (mg kg^{-1})*	*Hg (g kg^{-1})*	*Fe (g kg^{-1})*	*Mn (mg kg^{-1})*	*Al (g kg^{-1})*
Thale Noi (n = 14)										
mean	0.7	64.7	34.6	6.2	8.2	45.2	89.1	24.7	255	58.0
SD	0.3	4.5	4.4	1.8	1.7	5.0	14.9	4.7	124	9.5
range	0.4-1.4	59.0-76.0	23.4-40.5	2.9-9.2	5.7-10.8	36.0-55.2	63.4-113.1	18.0-34.1	133-548	39.4-78.9
Inner and Middle Lake (n = 37)										
mean	2.9	64.1	33.0	0.8	5.9	36.6	38.4	18.8	503	47.2
SD	5.2	9.1	6.9	0.4	1.5	9.2	11.1	5.4	292	12.7
range	0.5-32.4	30.4-74.6	22.5-50.4	0.2-2.1	3.7-10.8	23.7-73.3	24.3-68.0	12.6-42.5	168-1,339	28.9-76.2
Outer Lake (n = 23)										
mean	7.7	54.7	38.0	1.0	10.7	29.6	49.3	19.9	398	51.7
SD	17.9	16.1	7.8	0.4	5.5	9.2	15.2	5.4	146	14.2
range	0.7-66.3	4.8-71.5	21.9-57.9	0.5-2.0	5.1-25.7	7.9-48.2	26.6-110.3	7.2-34.7	139-776	17.6-71.4

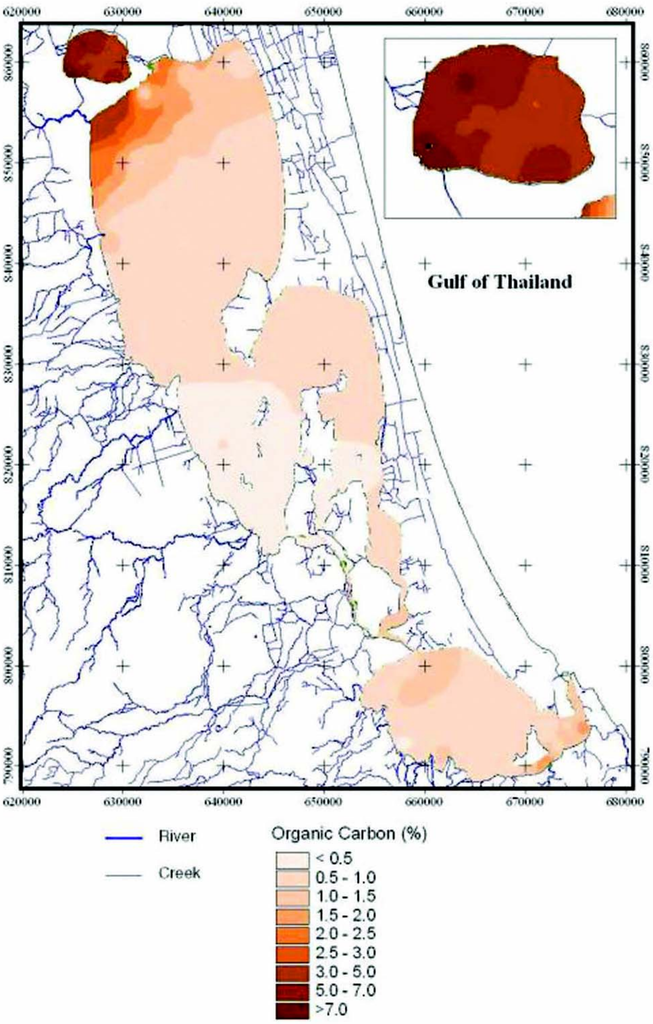

Figure 3: Spatial distribution of oxidizable organic carbon content in Songkhla Lake sediment.

The concentration of Cr is highest in Thale Noi sediment and decreases seaward. The range of Cr found in this study is ranged from 8 to 73 mg kg^{-1}, with the average of 45 ± 5, 37 ± 9 and 30 ± 9 mg kg^{-1} in Thale Noi, Inner & Middle Lakes and Outer Lake, respectively. Maneepong (1996) reported Cr concentration in Outer Lake at the range of 9 to 24 mg kg^{-1}. It is noted that Cr concentration in some area is almost two times higher than it was in the last decade. However, Cr concentration in the Songkhla Lake sediment is considered low compared to Tapi River (Suratthani, Thailand) and the Gulf of Thailand. Hungspreugs et al. (1989) reported the level of Cr in the unpolluted Tapi sediments at 46-97 mg kg^{-1}. Shazili et al. (1997 cited in Hungspreugs et al., 2003) reported the average Cr in the Gulf of Thailand sediments at 85 ± 15 mg kg^{-1} for September 1995 cruise and 63 ± 13 mg kg^{-1} for the April 1996 cruise. The Cr concentration in sediments from Lake Macquarie, New South Wales, Australia, was found in the range of 7 to 50 mg kg^{-1} (Roach, 2005).

Mercury concentration in Songkhla Lake sediments is found in the range of 24 to 113 µg kg^{-1}. Although Thale Noi is considered much less urbanized, its sediment contains Hg about two folds of those found in the main lake system (Table 1). This could be explained by the much higher OC in Thale Noi sediments than other parts. The Hg values found in Songkhla Lake is higher than those found in bottom sediment from remote mountain lake in the UK which is 20 µg kg^{-1} (Yang & Rose, 2003). In other parts of Thailand, Bangpakong River estuary sediment, Hg was found in the range of 120 to 480 µg kg^{-1} (Thongra-ar, 2001). Chongprasith and Wilairatanadilok (1999) reported the range of total Hg found in entire coast of the Gulf of Thailand and the Andaman Sea from 47 to 2,135 µg kg^{-1} with the average of 136 µg kg^{-1}. The Gulf of Thailand has high activity of natural gas exploration, which is an important source of Hg. The elevation of Hg concentration in Outer Lake comparing to Inner and Middle Lakes is probably due to inputs of effluents from Hat-Yai and Songkhla Municipalities. However, Hg content in Songkhla Lake surficial sediments is in the same range of those found in non-contaminated areas, e.g. Kara Sea (30 µg kg^{-1}), Ob estuary (35 µg kg^{-1}) and Yenisey estuary (50 µg kg^{-1}) in Russia (Loring et al., 1998), and Enid Lake (34 µg kg^{-1}) and Sardis Lake (31 µg kg^{-1}) in North Missisippi (Huggett et al., 2001). The value of Hg in clean ocean sediment reported by Paasivirta (1991) was 100-1000 µg kg^{-1}. Total Hg in sediment from Lake Macquarie, New South Wales, Australia, was found in the range of 10 to 2370 µg kg^{-1} (Roach, 2005).

Correlation of Arsenic, Chromium and Mercury with Dominant Metal Binding Components

Trace metals variability of sediments may be natural or influenced to some degree by anthropogenic sources. Since metals from such sources normally accumulate together mostly in the fine grained sediment fraction, examination of enrichment first requires compensation for the grain size and mineralogical effects on the metals variability in different samples. Normalization of As, Cr and Hg in sediments with dominant metal binding components is illustrated in Figure 4.

No correlation between As, Cr and Hg with percent clay size particles in Thale Noi sediments has been found. Clay size particles may play a role in controlling As and Hg distribution in the main lake system sediment. While

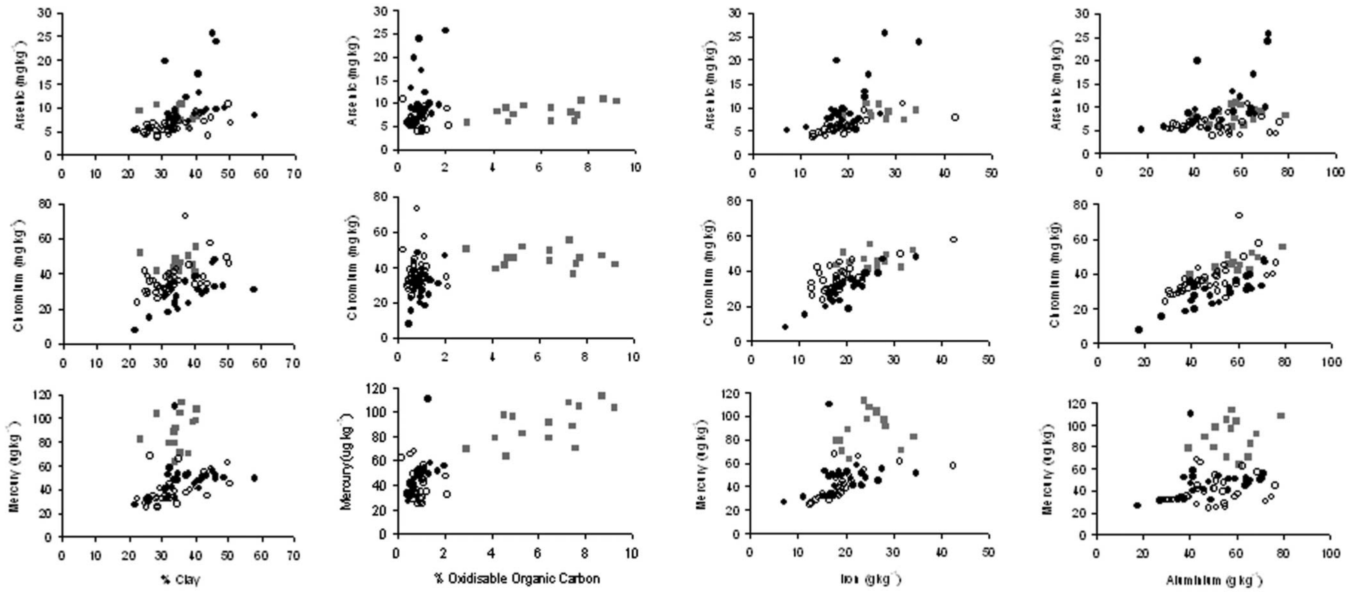

Figure 4: Relationship between aresenic, chromium and mercury with percent clay size particles, percent oxidizable organic carbon, iron and aluminium in sediments of Songkhla Lake (■ Thale Noi; ○ Inner and Middle Lakes; ● Outer Lake).

As and Cr has no correlation with OC content in sediments, a positive relationship with high correlation coefficients between Hg and OC, particularly in Thale Noi, suggested that Hg in sediments might be associated with OC. A positive correlation of Hg with OC could be expected in cases where Hg is primarily derived from wetland and soil (Wallschlager et al., 1996). Similar behaviour was found in sediments from Florida Bay (Kannan et al., 1998, Lawrence & Mason, 2001).

Chromium is strongly correlated with Fe and Al in sediments of the whole lake system (Figure 4). In Outer Lake sediments, As tends to correlate with Al (as well as clay fraction), exempt high concentration of four stations at the mouth of Pa-Wong canal while in Inner and Middle Lakes, As is associated with Fe.

Table 2: Loadings of the first three factors of arsenic, chromium, mercury, iron, manganese, aluminium, oxidisable organic carbon content and percent of materials <2 µm (clay size particles)

Components	*Loadings*		
	Factor 1	*Factor 2*	*Factor 3*
%clay size particles	-0.60	0.43	0.39
%OC	-0.64	-0.62	-0.29
As (mg kg^{-1})	-0.53	0.23	0.62
Cr (mg kg^{-1})	-0.82	0.04	-0.33
Hg (µg kg^{-1})	-0.73	-0.57	-0.01
Fe (g kg^{-1})	-0.84	0.01	0.11
Mn (mg kg^{-1})	-0.05	0.77	-0.52
Al (g kg^{-1})	-0.81	0.42	-0.21
% explained variance	45.3	21.4	13.2

Statistical Multivariate Analysis

The visual inspection of profiles for As, Cr and Hg with dominant metal binding components may not reveal a clear explanation of the geochemical factors controlling the dispersal and accumulation of the metals. In order to achieve this, interrelationship between variables and the associations of the metals with the main sedimentary and chemical components are presented by using varimax factor analyses as the indicators.

Principal component analysis was applied to the data matrices. The loading of the first three factors is presented in Table 2, and factor score plot of factors 1 and 2 is illustrated in Figure 5. The scattered scores plot shows two separated groups. This indicates the difference composition of the sediment samples in Thale Noi and the main lake.

The first three factors explain 79.9% of the total determinable variance. Factor 1 accounted for 45.3% of the variance. It contains significant loading on the Cr, Fe and Al and a lesser loading on Hg, %clay size particles and %OC. This indicates that Fe, Al, %clay size particles and %OC control the abundance of Cr and Hg in the sediments. However, %clay size particles and %OC has a lesser important role. Chromium and Hg vary directly with Fe and Al. This implies that Cr and Hg is somewhat

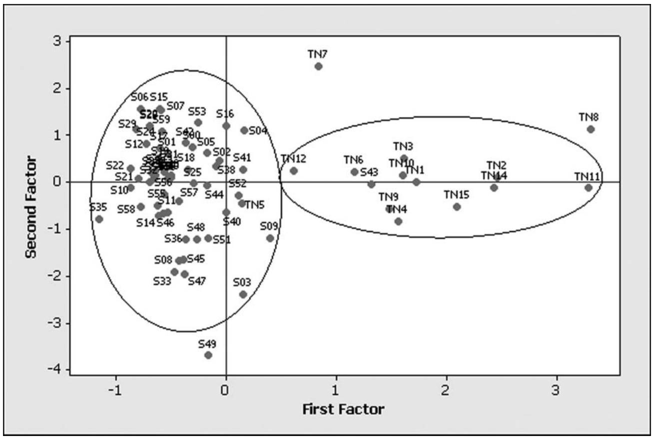

Figure 5: Score plot of factors 1 and 2 of surface sediment from Thale Noi (TN) and the main lake (S).

associated to land derived materials. However, fine grain sediment and organic matter also account to the accumulation of Cr and Hg in the sediment. Factor 2 (21.4% of the total variance) is Mn and %OC factor in which Mn decreases with increasing %OC. Factor 3 (13.2% of the total variance) has significant loading on As only.

Conclusion

In general, the surficial sediment of the Songkhla Lake system is considerably low in trace metal contents in all compartments of the lake system with the exception in Thale Noi for Hg and the mouth of Pa-Wong canal in the Outer Lake for As. The major geochemical factors that control the dispersal and accumulation of the metals in this lake system is iron oxides and aluminosilicate. Although organic matter and clay size particles seem to have less direct influence on the geochemistry of the metals, the factor loading is considerably significant. Therefore, the increase of organic matter in the lake system may change oxidizing/reducing environment and govern the biogeochemical processes of metals in the system.

Fast growing of economy in Songkhla Lake Basin may cause contamination problem from both organic and inorganic compounds. Currently, the lake has already been degraded through lack of understanding or concern for their carrying capacity. Although trace metals have not yet become the important pollutants in this region, knowledge about mechanism ruling metals behaviour in this lake system is inadequate. From the scientific point of view, metal cycling and biogeochemical modelling studies are of interests for further study.

Acknowledgements

The authors gratefully acknowledge the field and analytical assistance from our graduate students, Ms. Tittaya Sae-Eng, Mr. Jirawat Kaunkaew and Dr. Somkiat Korkiatwong. This research was funded by National Research Center for Environmental and Hazardous Waste Management (NRC-EHWM). Constructive comments on the manuscript from anonymous reviewers are greatly appreciated.

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