**Indonesian Journal of Environmental Science and Technology**
Vol.1 No.1, December 2014, pp.14-21
Submitted: August 27, 2014
Accepted: October 31, 2014

# Concentration, distribution, and correlation of heavy metals in seawater, sediment, and *Cerithidea obtusa* from coastal waters of Singkep Island, Riau Archipelago Province

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**Abstract:** Concentration, distribution and correlation of seven heavy metals in seawater, sediment and gastropod *Cerithidea obtusa* from coastal waters in the proximity of abandoned tin mining in Singkep Island has been evaluated. Samples were taken from five stations and heavy metal analysis was carried out by using AAS Perkin Elmer 3110 in Marine Chemistry Laboratory Faculty of Fisheries and Marine Science University of Riau. The mean concentrations of Cd, Cu, Pb, Zn, Ni, Mn and Cr were 0.0373, 0.0776, 0.1340, 0.1692, 01789, 0,1205 and 0.3232 mg/l in seawater; 2.0445, 11.5359, 24.6859, 23.7992, 13.2925, 13.1097 and 79.0821 µg/g in sediment and 3.03, 69.71, 14.30, 120.73, 11.62, 56.29 and 18.75 µg/g in *C. obtusa*, respectively. In general, the concentrations of metals in coastal water receiving effluents from abandoned tin mining activities in Singkep Island were higher than in coastal waters with less mining activities. Concentrations of all those metals in seawater were positively correlated with sediment and gastropod and its concentrations in sediment were also positively correlated with those in gastropod. Mean concentration of all metals in sediment were still below the ERM values, except for Cd which was exceeded the ERL but they were all still below the ERM values. Continuous environment monitoring programs should be implemented in Singkep Island coastal waters to evaluate possible increase of heavy metal threat as some new sand and bauxit mining activities are taking place.

**Key words:** heavy metal, seawater, sediment, *Cerithidea obtusa,* mining

## Introduction

Heavy metals in the marine environment will be dissolved in water and will accumulate in sediment and biota, and the concentrations may increase with time depending on the condition of the aquatic environment. Various human activities on land and in the sea may usually attributable to the pollution of coastal waters. The industrial and urban activities along the coastal areas may introduce a number of wastes containing heavy metals into the marine environ-ment which eventually cause environmental degradation and disruption of marine ecosystems [1,2,3,4,5,6].

Some research on heavy metals in seawater, sediment and biota in the coastal waters in Sumatra has been done [2,3,4,5,7,8,9,10,11,12,13]. However, in line with the development and increase human activity, the same types of research are still needed to be done, especially in the areas with a particular activity such as mining which may have negative impact on the environment and the health of the surrounding community.

Since long time ago, Singkep island has been used as mining areas for tin and bauxit by PT. Timah and some other private mining companies. Mining activities on the island has been conducted since 1887 by the Dutch Government through *Singkep Tin Maatschappij* under concessions from Billiton Maatschappij. In the last few years there have also been operating several private mining companies whose activities are more focused on the sand mining. Dangers which may arise from the mining activity is the accumulation of heavy metals by sediment, water and biota either directly or indirectly that will adversely affect the organisms. Among the organisms which able to accumulate heavy metals and could be used as excellent biomonitor for water pollution are gastropod snail *Cerithidea obtusa*.

Previous research in the coastal waters in the proximity of abandoned tin mining in Karimun Island showed the heavy metal concentrations were elevated significantly [14, 15]. Therefore, study of heavy metals in the coastal waters around Singkep island, especially in the surface water, sediment and snails *C. obtusa* would be able to provide information on the concentration, distribution and status of heavy metal contamination in these waters as well as baseline data for future relevant studies.

## Research Methods

This study was conducted in the coastal waters around Singkep island Riau Archipelago Province. Sampling of sea water, sediment and snails *C. obtusa* was conducted in March 2014 from five stations, namely Pengambil, Kuala Raya, Marok Tua, Marok Kecil and Sergang (**Fig** 1). Seawater samples were taken with 500 ml polyethylene sample bottles at 0-30 cm depth, the surface sediment samples (0-5 cm) were taken by using Eckman grab, while the *C. obtusa* samples were collected by hand from each station and they were then treated by following the procedures of Yap *et al.* [16,17] for sediment and snail and procedure of Hutagalung [18] for seawater analysis.

In the laboratory, sediment and soft tissue of snail samples were dried separately in an oven at temperature of 80-100 °C until a constant weight. Between 0.5 and 1.0 g of snail samples (soft tissues) and sediment (63 microns) were digested with $HNO_3$ for snails and in combination of $HNO_3$ and $HCIO_4$ for sediment (ratio 4:1), at low temperature (40°C) for 1 hour and then the temperature was increased to 140°C for 3 hours (Ismail and Ramli, 1997; Yap *et al.*, 2002). After the snails and sediment samples were completely digested, the solutions were cooled and diluted to 40 ml with double distilled water and filtered with No.1 whattman filter paper and stored in sample vials and then analyzed their heavy metals concentrations by AAS Perkin Elmer 3110 in Marine Chemistry Laboratory of Marine Science Department, Faculty of Fisheries and Marine Sciences University of Riau Pekanbaru. All statistical analyses for distribution and comparison of heavy metals in seawater, sediment and snails from each station were performed by using Statistical Package for Social Science (SPSS) version 17 [20].

## Results and Discussion

### Water Quality Parameters

The results of water quality parameter measurements in seawater (**Table1**) showed no significant difference of all parameters between stations. The pH value of 6-7 can be considered as still in the range of tolerable level by aquatic biota, while the temperature were also in the range for the life of the marine organism of 25-32$^0$C [21]. Nybakken [22] stated that the water temperature will determine the biological activity and their activeness on marine biota in the water. When compared with the water quality standards for marine biota [23], the average environmental parameters in coastal waters of Singkep island are generally still in the range of tolerable level for marine organisms.

### Concentrations of heavy metals in seawater

The highest metal concentration in sea water was found in Pengambil for Cd, Cu, Pb, Zn, Mn and Cr, whilst Ni was highest in Marok Tua. The lowest concentration for all metals were found in Sergang, except for Mn which was found in Kuala Raya.


**Fig 1.** Map of Singkep Island and sampling sites

**Table1.** Results of water quality parameter measurement

| Station | Temperature ($^0$C) | pH | Salinity (‰) | Transparency (m) | Current velocity (m/s) |
|---|---|---|---|---|---|
| 1 (Pengambil) | 30 | 6 | 20 | 0.75 | 0.60 |
| 2 (Kuala Raya) | 29 | 6 | 19 | 0.65 | 0.50 |
| 3 (Marok Tua) | 27 | 7 | 18 | 0.70 | 0.55 |
| 4 (Marok Kecil) | 28 | 6 | 18 | 0.75 | 0.65 |
| 5 (Sergang) | 29 | 7 | 20 | 0.80 | 0.70 |

**Table2.** Mean concentrations of heavy metals in seawater

| Station | Metal concentration (mg/l) | | | | | | |
|---|---|---|---|---|---|---|---|
| | Cd | Cu | Pb | Zn | Ni | Mn | Cr |
| 1 (Pengambil) | 0.0424 | 0.1288 | 0.3140 | 0.2389 | 0.1883 | 0.2714 | 0.7471 |
| 2 (Kuala Raya) | 0.0371 | 0.0675 | 0.1208 | 0.1927 | 0.1584 | 0.0698 | 0.2139 |
| 3 (Marok Tua) | 0.0385 | 0.0815 | 0.1378 | 0.1986 | 0.2394 | 0.0926 | 0.2624 |
| 4 (Marok Kecil) | 0.0349 | 0.0644 | 0.0513 | 0.1259 | 0.1611 | 0.0923 | 0.2034 |
| 5 (Sergang) | 0.0336 | 0.0458 | 0.0462 | 0.0897 | 0.1470 | 0.0764 | 0.1894 |

The results of the Anova test showed significant difference ($p<0.05$), except for Cd ($p>0.05$) between station for the concentration of heavy metals in sea water and therefore followed by Tukey HSD test to see the comparison between the station. There are significant differences ($p <0.05$) for Cu, Pb, Zn,Ni, Mn and Cr between the stations, except for Cu (Station 2- 3 and 2-4), Pb (Station 2-3 and 4-5), Zn (Station 1-3, 2-3, and 4-5), Ni (Station 1-2, 1-4, 2-4, 2-5, and 4-5), Mn and Cr (Station 2-3, 2-4, 2-5, 3-4, 3-5, and 4-5).

Station 1 was found to have the highest concentration for most of the metals analyzed and the lowest concentrations for those metals were in Station 5. This is presumably due to Station 1 is an area receiving run off from abandoned mining area through river and from tailing that have been left around the sampling location and some newly constructed bauxit mining wastes, while Station 5 was a recreational area that is relatively not affected by the mining activities.

The mean concentrations of metals in seawater samples from the mining areas (0.0382; 0.0855; 0.1560; 0.1890; 0.1868; 0.1315; and 0.3567 mg/l) were higher than that in the recreational area (0.0336; 0.0458; 0.0462; 0.0897; 0.1470; 0.0764; and 0.1894 mg/l) as can be seen in **Fig 2**. Increse metal concentrations in sediment in a neraby mining areas has also been reported [24,25,26].

Seawaters are naturally contain heavy metals in small amounts, but when the concentration in the water increases due to certain sources, the amount can be toxic to the organisms or humans who consume them. In general, all heavy metals can cause negative effect to aquatic organisms at certain concentration limits. The influence varies depending on the type of metal, the species of organisms, the permeability and the detoxication mechanism [27]. Heavy metals in the water easily absorbed and accumulate in phyto-plankton which is the starting point of the food chain, further higher up through the food chain to other organisms [28]. Heavy metal concentration in the waters is always changing depending on the time of disposal of waste and the degree of aquatic environment perfection. Increased concentrations of heavy metals in seawater will be followed by an increase in heavy metals in the body of organisms, so that sea water pollution will be followed by the biota in the environment.

**Concentrations of heavy metals in Sediment**

Average concentration of analyzed metals in the sediment at each station can be seen in **Table 3**. Metal concentrations were found to be highest in Station 3 and the lowest was in stations 5. The results of comparison between stations indicated that significant different ($p<0.05$) was found for Cd, whilst other metals showed highly significant different between stations ($p<0.01$).

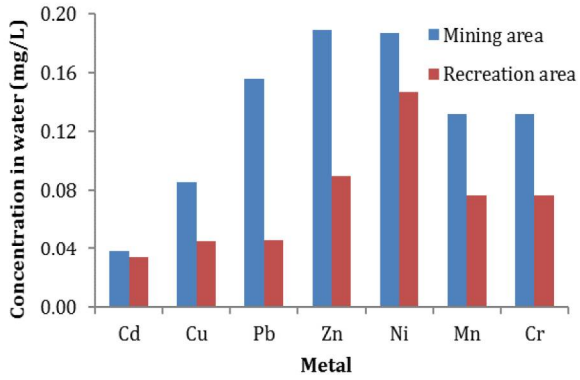


**Fig 2.** Comparison between metal concentrations in seawater from mining and recreation area

Once heavy metals enter the seawater it will experience precipitation, dilution and dispersion. Heavy metal that settles on the sea floor will be accumulated into the sediments [29]. As in sea water, the concentrations of metals in the sediment at Station 3 as an area of more mining activities also has the highest concentration of all metals and it was assumed that the metals were derived from these abandoned and active mining activities further up in the nearby streams. There are many piles of mining wastes were left exposed at several places close to the huge hole or man-made lake as a result of mining activities in the past. The high concentration of metals in the region is also thought to come from other anthropogenic activities. Marok Tua, Marok Kecil and Pengambil coastal waters were also used as human settlement, housing and most of fishermen boats are always mooring in this area. As also for seawater. the mean concentrations of metals in sediment samples from mining areas was higher than that in the recreation area as can be seen in **Fig 3**. Sandy sediment in Station 5 was also assumed to be the cause of lower metal concentrations. Owen and Shandu [30] stated that sandy sediment would accumulate relatively lower metal concentration in comparison to muddy sediments.

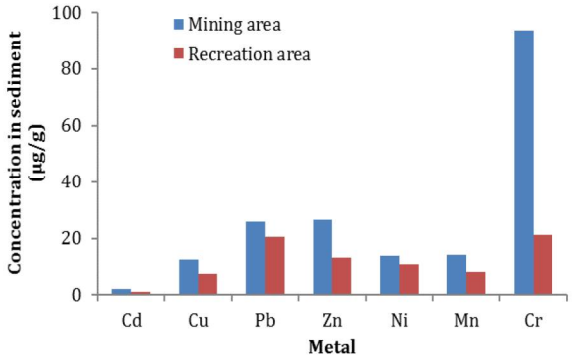


**Fig 3.** Comparison between metal concentrations in sediment from mining and recreation area

**Table 3.** Mean concentrations of heavy metals in sediment

| Station | Metal concentration (µg/g) | | | | | | |
|---|---|---|---|---|---|---|---|
| | Cd | Cu | Pb | Zn | Ni | Mn | Cr |
| 1 (Pengambil) | 2.3740 | 11.1550 | 25.4740 | 16.8960 | 13.2365 | 14.5059 | 86.8923 |
| 2 (Kuala Raya) | 2.0540 | 9.6260 | 21.0260 | 26.1920 | 13.1828 | 12.3896 | 94.2380 |
| 3 (Marok Tua) | 2.5220 | 20.9970 | 32.8593 | 48.8890 | 16.5970 | 19.1238 | 136.6763 |
| 4 (Marok Kecil) | 1.9580 | 8.3290 | 23.5800 | 14.0130 | 12.6026 | 11.2140 | 56.4643 |
| 5 (Sergang) | 1.3144 | 7.5725 | 20.4900 | 13.0060 | 10.8436 | 8.3149 | 21.1397 |

**Table 4.** Mean concentrations of heavy metals in *C. obtusa*

| Station | Metal concentration (µg/g) | | | | | | |
|---|---|---|---|---|---|---|---|
| | Cd | Cu | Pb | Zn | Ni | Mn | Cr |
| 1 (Pengambil) | 3.2471 | 73.3663 | 14.6382 | 126.5533 | 11.8276 | 67.7247 | 21.2460 |
| 2 (Kuala Raya) | 2.8521 | 71.4793 | 14.1984 | 117.1343 | 12.1709 | 56.9065 | 18.7877 |
| 3 (Marok Tua) | 3.5353 | 78.1950 | 19.2163 | 144.5903 | 13.1315 | 74.8675 | 23.8942 |
| 4 (Marok Kecil) | 2.8634 | 68.6023 | 12.5552 | 115.2913 | 10.9961 | 49.6389 | 16.3064 |
| 5 (Sergang) | 2.6543 | 56.9171 | 10.8713 | 100.0590 | 9.9979 | 32.3308 | 13.5374 |

### Concentration of metals in snail *C. obtusa*

The mean concentration of metals in snails *C. obtusa* from coastal waters of Singkep Island (**Table 4)** had the same distribution profile with the concentrations of metals in seawater and sediment. Station 3 showed the highest concentration compared to the other stations and the lowest was at Station 5 as recreation area. The order of metal concentrations in snail was Zn>Cu>Mn>Cr>Pb> Ni>Cd.

The mean concentrations of metals in *C. obtusa* samples from mining areas were higher than those

in the area with relatively smaller number of tin mining activities in the recreation area (3.1263; 72.9125; 151.5225; 125.8943; 12.0322; 62.2857; 20.0592; 2.6567; 87.2733; 108.7140; 100.0613; 9.9980; 32.3327; and 13.5380 µg/g) for Cd, Cu, Pb, Zn, Ni, Mn, and Cr respectively (**Fig 4**).

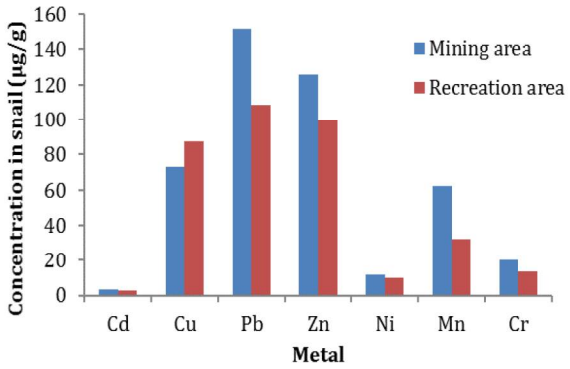


**Fig 4.** Comparison between metal concentrations in *C. obtusa* from mining and recreation area

## Relationship Between Metal Concentrations in Water, Sediment and Snails *C. obtusa*

Linear regression analysis clearly showed positive correlations (r= 0.182-0.844) while relationship between metal concentrations in sediment and snail had a stronger correlation coefficient (0.720) compared to metal concentration in water and snail (0.486) and between concentration in water and sediment (0.332). Relationship between metals concentration in the seawater with metal concentrations in the sediment and snails *C. obtusa* as well as the relationship between the concentration of metals in snails and sediments from coastal waters of Singkep Island can be seen in **Table 5**.

## Coastal Water Pollution Status

To determine the level of pollution in the coastal waters of Singkep Island, the concentration of heavy metals in the sediment were compared to ERL (Effects Range Low) and ERM (Effects Range Median) standards as proposed by Long *et al.* [31, 32]. Comparison of metal concentrations during the study with the standard quality guidelines can be seen in **Table 6**.

Concentrations of Cd in all stations and Cr in Station 1,2, and 3 were all exceeded the ERL values but still below the ERM values which means that the metals concentrations in these stations might have negative impact on organisms that exist in those waters.

**Table 5.** The relationship between concentration of metals in water, sediment, and snails *C. obtusa*

| Relationships | Regression Equations | $R^2$ | r |
|---|---|---|---|
| Metal water - sediment | $y_{Cd} = 0.15 + 50.88x$ | 0.214 | 0.463 |
| | $y_{Cu} = 7.83 + 47.71x$ | 0.073 | 0.270 |
| | $y_{Pb} = 23.09 + 11.86x$ | 0.046 | 0.214 |
| | $y_{Zn} = 8.17 + 92.36x$ | 0.172 | 0.416 |
| | $y_{Ni} = 7.21 + 34.02x$ | 0.288 | 0.537 |
| | $y_{Mn} = 11.67 + 11.91x$ | 0.059 | 0.243 |
| | $y_{Cr} = 68.44 + 32.90x$ | 0.033 | 0.182 |
| Metal water - snail | $y_{Cd} = 2.34 + 18.40x$ | 0.082 | 0.286 |
| | $y_{Cu} = 58.28 + 147.2x$ | 0.353 | 0.594 |
| | $y_{Pb} = 12.84 + 10.82x$ | 0.141 | 0.375 |
| | $y_{Zn} = 94.13 + 157.1x$ | 0.411 | 0.641 |
| | $y_{Ni} = 7.42 + 23.48x$ | 0.550 | 0.742 |
| | $y_{Mn} = 46.98 + 77.27x$ | 0.160 | 0.400 |
| | $y_{Cr} = 16.65 + 6.49x$ | 0.131 | 0.362 |
| Metal sedimen - snail | $y_{Cd} = 2.42 + 0.30x$ | 0.260 | 0.510 |
| | $y_{Cu} = 58.68 + 0.95x$ | 0.463 | 0.680 |
| | $y_{Pb} = 4.75 + 0.39x$ | 0.545 | 0.738 |
| | $y_{Zn} = 99.91 + 0.87x$ | 0.629 | 0.793 |
| | $y_{Ni} = 7.32 + 0.32x$ | 0.421 | 0.649 |
| | $y_{Mn} = 12.73 + 3.32x$ | 0.713 | 0.844 |
| | $y_{Cr} = 12.30 + 0.08x$ | 0.681 | 0.825 |

**Table 6.** Comparison of heavy metal concentrations in the sediments with Standard ERL and ERM

| Metal | Concentration (µg/g) | | | | | | |
|---|---|---|---|---|---|---|---|
| | St.1 | St.2 | St.3 | St.4 | St.5 | ERL* | ERM* |
| Cd | 2.37 | 2.05 | 2.52 | 1.96 | 1.31 | 1.20 | 9.60 |
| Cu | 11.16 | 9.63 | 21.00 | 8.33 | 7.57 | 34.00 | 270.00 |
| Pb | 25.47 | 21.03 | 32.86 | 23.58 | 20.49 | 46.00 | 218.00 |
| Zn | 16.90 | 26.19 | 48.89 | 14.01 | 13.01 | 150.00 | 410.00 |
| Ni | 13.24 | 13.18 | 16.60 | 12.60 | 10.84 | 20.90 | 51.60 |
| Cr | 86.89 | 94.24 | 136.68 | 56.46 | 21.14 | 81.00 | 370.00 |

* Long *et al* [31,32]

Other metals in all stations were still below both ERL and ERM values. If the value of the concentration of heavy metals was passed ERL values but remained below the ERM value means that there could be negative effects on aquatic organisms, and when the concentration of heavy metals exceeded the ERM value then the concen-tration of heavy metals have negative effect on aquatic organisms [31,32]. Higher concentrations of all metals analyzed in the mining areas compared to relatively non-mining area in Singkep island was due to the difference in the condition of the coastal water environment. The influence of tailing and other related activities provide significant input of heavy metals to coastal waters of Marok Tua, Marok Kecil, Pengambil and Kuala Raya when compared to Sergang coastal waters.

## Conclusion and recommendations

Heavy metal concentrations in Singkep island coastal waters were found to be different for each station, where the concentrations in sea water, sediment and the snail *C. obtusa* in the proximity of mining areas were higher than in station nearby recreation area. In general, concentration of heavy metals in sea water with sediment and metal concentration in sediments with concentration in snails has positive relationships. Based on Sediment Standard Quality Guideline of ERL and ERM the concentrations of metals in coastal waters around Singkep island might have negative impacts on organisms that living in those waters. Therefore, continuous monitoring of coastal waters around Singkep island is needed to avoid possible negative impacts on the quality of the coastal environment.

## Acknowledgement

The authors thank to Directorate General Higher Education Ministry of Education and Culture which has provided financial support through Fundamental Research Scheme Fiscal Year 2014.

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