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Bioaccumulation of Heavy Metals in Aquatic Animals Collected from Coastal Waters of Gresik, Indonesia

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Abstract: A survey of the presence of metals in aquatic animals caught from Gresik coastal waters, Indonesia, has been conducted. The results show that all animals contained Zn levels higher than the level of other metals (As, Cd, Cr, Ni, Cu, Se and Pb) in the same species. The copper level recorded in tissue of banana shrimp showed higher copper concentration than that in other fishes. The concentration of Zn in whole body of animals (ponyfish and anchovy) was relatively higher than those recorded in muscle tissues (banana shrimp, drum, mullet and sea catfish). It was also found that the level of metals in all samples collected from this area contained metals in their tissue within acceptable range for consumption. These data provide useful information for future reference.

Key words: Metals, shrimp, fish, bioaccumulation, coastal waters.

Introduction

Heavy metals naturally occur in seawater in very low concentrations, but their concentration levels have increased due to anthropogenic pollutants over time (Kargin et al., 2001). Industrial activities as well as agriculture and mining create a potential source of heavy metals pollution in aquatic environment (Unlu et al., 1996). Pollution of aquatic ecosystems by heavy metals is an important environmental problem, as heavy metals constitute some of the most dangerous toxicants that can bioaccumulate (Soegianto et al., 1999a; Benson et al., 2007). Metals that are deposited in the aquatic environment may accumulate in the food chain and cause ecological damage also posing a threat to human health due to biomagnification over time (Yilmaz and Yilmaz, 2007). Aquatic organisms have been reported to accumulate heavy metals in their tissues several times above ambient levels (Canli and Atli, 2003). These metal levels in aquatic animals should be monitored regularly to check animal health and in view of the quality of public food supplies.

Gresik is one of the big industrial cities in Indonesia. Industrialization and urbanization have proceeded rapidly during past two decades in this city, including the development of large harbours and heavy industries, resulting in the deterioration of neighbouring marine environment. The coastal waters of Gresik is believed to receive wastewater discharges from number of wastewater treatment facilities of industries located along Gresik coastal zone. The potential industries which contribute to the level of metals in these coastal waters are a superphosphate plant, an asphalt plant, a coal-fired electric power plant, metal smelters and refineries, natural gas processing plants, etc. These waters represent also the habitat of some edible organisms caught by local fishermen. Many people still use the coastal waters as their fishing ground. This study was undertaken to measure the heavy metal contamination in the tissues of aquatic animals collected from Gresik coastal waters of East Java. The data are then compared with the standard

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for maximum limits of metals in the tissue of marine biota.

Material and Methods

Five species of fishes, ponyfish (*Leiognathus equlus*), anchovy (*Coilia dusumieri*), drum (*Johnius belengeri*), mullet (*Mugil vaigiensis*) and sea catfish (*Arius leptonotacanthus*), and one species of shrimp (*Penaeus merguensis*), were chosen as samples for metal analyses. The species selected for metal analyses was based on general abundance in the area, similarity of their size and their potential to be consumed by local people. The animals were collected by gillnets in Gresik coastal waters in June 2005 (Figure 1).

Depending on availability, a number of animals of each species were processed for metal analysis. The samples were homogenized prior to metal analysis. Fish species where entire body is consumed such as anchovy and ponyfish, samples of the whole body were homogenized, and prepared for metals detection. For other fishes and shrimp, only edible part or flesh were used for metal detection. Before filleted, external water of each individual sample was absorbed using tissue papers. The flesh then weighed to the nearest 0.1 g on an analytical balance, minced by knife and added to a known amount of double de-ionized water, then pureed using a multi-speed blender. The anchovy and ponyfish were also undergoing the same process as well as other species. Each sample then was put into special flask separately, weighed, then frozen at –20°C for a period not less than eight hours. The frozen sample was then placed under vacuum on a freeze-dryer unit (Labconco) until sample was completely dried. Dried sample was weighed and approximately 1 g of each was digested in 5 ml of high purity HNO_3 and 5 ml of high purity HCl using Microwave Digester (Ethos D) for approximately 25 minutes. Digests are filtered through Whatman 44 ashless filter paper and made up to 50 ml with double de-ionized water.

Inductively-coupled plasma emission spectroscopy (Thermo Jarrell Ash Type: IRIS Advantage) was used

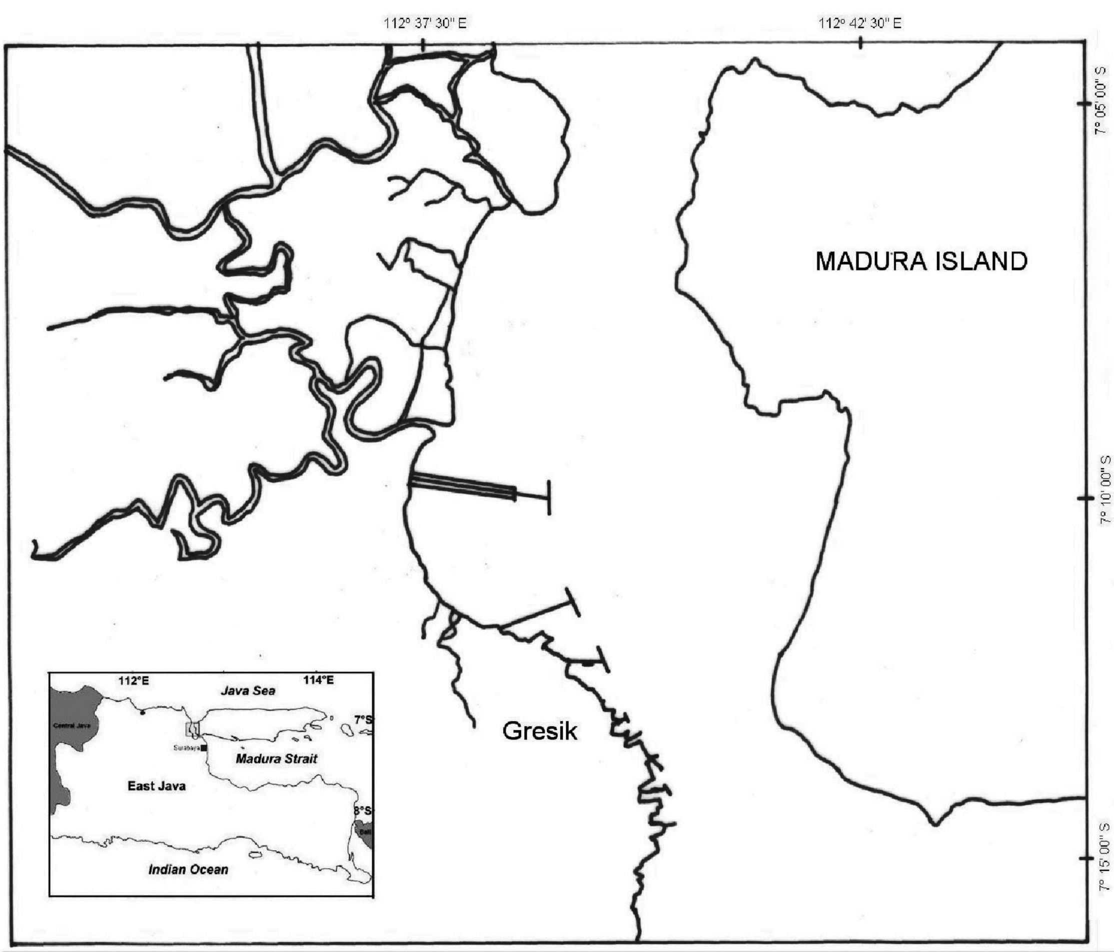

Figure 1: Sampling location of aquatic animals.

for determination of arsenic (As), cadmium (Cd), copper (Cu), zinc (Zn), lead (Pb), chromium (Cr) and nickel (Ni). Analytical blanks were run in the same way as the samples and concentrations were determined using standard solutions prepared in the same acid matrix. Accuracy and precision of the results were checked and compared with standard reference material (dogfish muscle reference materials, DORM-2). The standard reference material digests were found to conform with the documented values for certified trace metal concentrations. All metal concentrations were quoted as mg.kg^{-1} dry weight.

Results

The concentration of metals in aquatic animals is presented in Table 1. The data show that all animals caught from Gresik coastal waters, Indonesia, contained Zn levels higher than the level of other metals (As, Cd, Cr, Ni, Cu, Se and Pb) in the same species. The copper level recorded in tissues of banana shrimp showed higher copper concentration than that in other fishes. Our findings also showed that the concentration of Zn in whole body of animals (ponyfish and anchovy) were relatively higher than that recorded in muscle tissues (banana shrimp, drum, mullet and sea catfish).

Discussion

A wide range of values for various metals was found in different species of marine biota, as well as in flesh and whole body samples. The concentration of zinc in both fishes and shrimp were relatively higher compared to concentration of other metals in the same animals. The similar findings were also recorded in fishes (Parsons, 1999; Miramand et al., 2001; Zehra et al., 2003; Tyrrell et al., 2005) and crustaceans (Ridout et al., 1985; Swaileh and Adelung, 1995; Parsons, 1998; Hossain and Khan, 2001; Miramand et al., 2001; Tyrrell et al., 2005) caught from other waters of the world. The copper level recorded in tissue of shrimp samples (*Penaeus merguensis*) showed elevated copper concentration than that in other fishes. This level is presumably influenced by the copper contained in the haemolymph of crustaceans. Both copper and zinc are essential elements and their concentrations are usually regulated by marine fishes (Thompson, 1990) and crustaceans (White and Rainbow, 1982; Rainbow, 1995; Soegianto et al., 1999b).

Almost all metals measured in this study are relatively lower than the values recorded in aquatic animals from other regions of the world. The findings of other studies are summarized in Table 2, and are compared with the concentrations reported in this study and elsewhere in the world.

The levels of heavy metal in aquatic animals vary in various species and different aquatic environments (Canli and Atli, 2003). As reported in our study, trace element concentrations varied markedly among species. These variations are presumably due to individual samples being of different size categories, from different ecological niches, and from different trophic levels. Possibly, species also have different metabolic requirements for specific trace element. Bottom-dwelling and demersal species usually had higher concentrations than more pelagic species, which may be related to greater exposure to

Table 1: Heavy metal concentrations measured in tissue samples (mg kg^{-1} dry weight) of aquatic animals collected from coastal waters of Gresik in June 2005

Contaminant	*Species*											
	Banana shrimp		*Ponyfish*		*Anchovy*		*Mullet*		*Sea Catfish*		*Drum*	
	N = 26		*N = 20*		*N = 18*		*N = 4*		*N = 5*		*N = 6*	
	W = 9.8 ± 1.1		*W = 10.7± 1.3*		*W = 6.4 ± 0.8*		*W = 19.3 ± 2.4*		*W = 19.8 ± 2.7*		*W = 13.8 ± 2.0*	
	Mean	*SD*	*Mean*	*SD*	*Mean*	*SD*	*Mean*	*SD*	*Mean*	*SD*	*Mean*	*SD*
Copper	1.166	0.162	0.205	0.085	0.049	0.027	0.108	0.032	0.098	0.011	0.039	0.025
Lead	0.022	0.008	0.039	0.016	0.028	0.013	0.047	0.002	0.040	0.036	0.021	0.013
Zinc	2.132	0.280	2.264	0.898	2.437	0.545	0.435	0.078	1.035	0.038	0.482	0.046
Arsenic	0.084	0.015	0.003	0.001	0.014	0.007	0.013	0.008	0.027	0.021	0.048	0.046
Selenium	0.014	0.014	0.026	0.020	0.018	0.018	<0.001	<0.001	0.020	0.016	0.014	0.008
Cadmium	0.0002	0.0001	0.0012	0.0010	0.0013	0.0010	<0.0001	<0.0001	0.0006	0.0004	0.0004	0.0003
Nickel	0.008	0.005	0.004	0.003	0.027	0.008	0.012	0.010	0.005	0.001	0.007	0.005
Chromium	0.002	0.001	0.016	0.005	0.056	0.019	0.008	0.006	0.009	0.001	0.003	0.002

Note: N = number of animals; W = weight (gram); SD = standard deviation

Table 2: A comparison of heavy metal concentrations (mg kg^{-1} dry weight) in aquatic animals collected from Gresik coastal waters and other regions of the world

Location	*Species*	*As*	*Cd*	*Cr*	*Cu*	*Ni*	*Pb*	*Se*	*Zn*	*References*
Gresik coastal waters, Indonesia	*Penaeus merguensis* (Crustacea, decapod)	0.084 ± 0.015	0.0002 ± 0.0001	0.002 ± 0.001	1.166 ± 0.162	0.008 ± 0.005	0.022 ± 0.008	0.014 ± 0.014	2.132 ± 0.280	Present study
	Leiognathus equulus (Teleostei)	0.003 ± 0.001	0.0012 ± 0.0010	0.016 ± 0.005	0.205 ± 0.085	0.004 ± 0.003	0.039 ± 0.016	0.026 ± 0.020	2.264 ± 0.898	Present study
	Coilia dusumieri (Teleostei)	0.014 ± 0.007	0.0013 ± 0.0010	0.056 ± 0.019	0.049 ± 0.027	0.027 ± 0.008	0.028 ± 0.013	0.018 ± 0.018	2.437 ± 0.545	Present study
	Johnius belengeri (Teleostei)	0.048 ± 0.046	0.0004 ± 0.0003	0.003 ± 0.002	0.039 ± 0.025	0.007 ± 0.005	0.021 ± 0.013	0.014 ± 0.008	0.482 ± 0.046	Present study
	Mugil vaigiensis (Teleostei)	0.013 ± 0.008	<0.0001	0.008 ± 0.006	0.108 ± 0.032	0.012 ± 0.010	0.047 ± 0.002	<0.001	0.435 ± 0.078	Present study
	Arius leptonotacanthus (Teleostei)	0.027 ± 0.021	0.0006 ± 0.0004	0.009 ± 0.001	0.098 ± 0.011	0.005 ± 0.001	0.040 ± 0.036	0.020 ± 0.016	1.035 ± 0.038	Present study
Hongkong	Natantian decapods	-	<0.9-59.29	<0.9-13.95	8.91-113.1	<0.9-65.40	<0.9-248.4	-	9.05-62.77	Parsons (1998)
	Johnius belengeri (Teleostei)	<0.9-96	<0.9-13.5	<0.9-3.5	<0.9-17.2	<0.9-6.6	<0.9-60	<0.9-36.5	10-50	Parsons (1999)
	Leiognathus brevirostris (Teleostei)	<0.9-59.8	<0.9-9.7	<0.9-3.8	<0.9-8.2	<0.9-5	<0.9-102	<0.9-28.1	30.5-63.5	Parsons (1999)
Bengal bay, Bangladesh	*Penaeus monodon* (Crustacea decapod)	-	0.2-0.3	1.7-2.9	12.2-21.3	2.9-5.9	0.8-1.3	-	24.2-35.7	Hossain and Khan (2001)
	Panulirus polyphagus (Crustacea decapod)	-	0.3-0.4	2.5-3.1	25.8-35.7	3.1-7.0	1.0-1.9	-	17.6-64.5	Hossain and Khan (2001)
East Atlantic Ocean	*Systellapsis debilis* (Crustacea decapod)	-	4.3-5.7	-	19-176	-	-	-	38-177	Ridout et al. (1985)
Western Baltic	*Diastylis rathkei* (Crustacea: Cumacea)	-	0.17-0.56	-	61.7-172.4	-	2.9-14.8	-	54.3-120.3	Swaileh and Adelung (1995)
Irish Ports	*Nephrophs norvegicus* (Crustacea decapod)	-	0.28	<0.28	17.4	-	<0.24	-	53.2	Tyrrell et al. (2005)
	Scomber scombrus (Teleostei)	-	<0.016	<0.28	2.12	-	<0.24	-	14.96	Tyrrell et al. (2005)
	Pleuronectes platessa (Teleostei)	-	<0.016	<0.28	<0.64	-	<0.24	-	17.28	Tyrrell et al. (2005)
Baluchistan Coast, Pakistan	*Acanthopagurus berda* (Teleostei)	-	0.04-0.11	-	0.30-0.55	-	0.25-0.50	-	3.65-4.32	Zehra et al. (2003)
Seine Estuary	*Dicentrarchus labrax* (Teleostei)	-	0.016-0.035	-	2.7-42	-	0.12-0.26	-	54-79	Miramand et al. (2001)
	Platichthys flesus (Teleostei)	-	0.019-0.052	-	2.8-3.4	-	0.34-0.92	-	72-192	Miramand et al. (2001)
	Crangon crangon (Crustacea decapod)	-	0.08-014	-	48.5-81	-	0.4-0.9	-	57-105	Miramand et al. (2001)
	Palaemon longirostris (Crustacea decapod)	-	0.05-0.23	-	67.5-1007	-	0.3-1.0	-	79-89	Miramand et al. (2001)
Gulf of Cambay, India	*Harpodon nehereus* (Teleostei)	1.74 ± 0.865	0.23 ± 0.029	0.77 ± 0.054	2.37 ± 0.451	ND	1.09 ± 0.071	-	38.24 ± 1.641	Reddy et al. (2007)
	Metopograpsus maculates (Crustacea decapod)	ND	1.6 ± 0.566	2.075 ± 0.389	175.45± 2.45	3.15 ± 0.041	2.775 ± 0.177	-	44.22 ± 1.21	Reddy et al. (2007)

Note: ND = nondetectable, - = No data

Table 3: Maximum residue limit and maximum permitted concentration of metals in marine biota muscle (mg kg^{-1} dry weight) from various countries and organizations

No.	*Contaminant*	*U.K.*[1]	*Australia*[2]	*Hong Kong*[1]	*European Regulation 466/2001/EC*[3]	*Indonesia*[4]	*IRPTC*[5]
1	Cadmium	-	0.80	8	0.4	-	-
2	Lead	-	6.0	24	1.6	8	-
3	Chromium	-	-	4	-	-	-
4	Copper	80	-	-	-	80	-
5	Zinc	200	-	-	-	400	-
6	Arsenic	-	4^+	-	-	4	-
7	Nickel	-	-	-	-	-	2
8	Selenium	-	4	-	-	-	-

Note: 1. Parsons (1998, 1999); 2. Otway (1992); 3. Tyrrell et al. (2005); 4. Decree of General Director of Food and Drug Supervision No. 03725/B/SK/VII/89 concerning maximum limit of metals in food; 5. IRPTC (1988); + Inorganic arsenic, this study measured total arsenic.

contaminated sediments (Trucco et al., 1990; Parsons, 1999). Kargin (1996) stated that due to variations in feeding habits, habitat and behaviour of species, the level of metals found in tissues of the demersal species were always higher than those found in pelagic species. The results presented in this study showed that concentration of some metals (Zn, Cd, Cr and Se) in catfish (demersal species) were relatively higher than that recorded in muscle tissues of pelagic fishes (mullet and drum).

The potential toxicological impacts of contaminated seafoods can be evaluated on the basis of concentrations in whole body and flesh samples. Concentrations of trace elements can be 200-400% greater in organs and other tissues than in muscle (Thompson, 1990; Chan, 1995). Thus, one might expect higher concentrations to be recorded in homogenized whole body samples. In our study, the concentrations of certain metals (Zn, Cr and Cd) in whole body of fish (anchovies and ponyfish) were relatively higher than those documented in muscle tissue of mullet, drum and sea catfish. This difference is presumably due to higher concentration of metals in the viscera of animal. Target organs, such as liver, gonads, kidney and gills, have a tendency to accumulate heavy metals in high values (Yilmaz, 2005). It is generally accepted that muscle is not an organ in which metals accumulate (Legorburu et al., 1988). Similar results were reported from a number of fish species showing that muscle is not an active tissue in accumulating heavy metals (Karadede and Unlu, 2000; Kargin and Erdem, 1991).

Comparing the present data with guidelines and limits (Table 3), it can be seen that most of metal concentrations found in the tissues of aquatic animals proved to be below the tolerance levels for human consumption.

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