Asian Fisheries Science **21**(2008):395-404
https://doi.org/10.33997/j.afs.2008.21.4.004

Asian Fisheries Society, Manila, Philippines

Trace Metal Concentrations in Aquatic Animals Collected from the Coastal Waters of Gresik, Indonesia

A. SOEGIANTO[1*], B. IRAWAN[1] and HAMAMI[2]

[1]Department of Biology
[2]Department of Chemistry
Airlangga University
Kampus C, Jl. Mulyorejo, Surabaya
60115 Indonesia

Abstract

A survey on the presence of metals in five species of fish and one species of shrimp was conducted in 2003 and 2004 along the Gresik coastal waters, Indonesia. The results showed that almost all metals measured in this study are relatively lower than the values recorded in aquatic animals from other regions of the world. Only the level of zinc was comparable to the results of the other researchers. It was also found that the level of metal in all samples collected from this area contained metal in their tissue within acceptable range for consumption. These data provide useful information for future reference.

Introduction

One aspect of environmental degradation is pollution from metals which are persistent and are bioaccumulated by aquatic organism, with serious public health implications (Rainbow 1993). Evidence of long-term adverse effect such as cancer, due to metals in marine animals comes from a number of field and experimental studies (Mackey et al. 1996; Zauke et

[*] Corresponding author. Tel.: +62 31 593 6501, Fax. +62 31 593 6502
E-mail address: agoes_soegianto@unair.ac.id

al. 1999). Several studies have revealed that metal is also dangerous to aquatic organisms and that it can be bioaccumulated in the food chain (Rainbow 1995; Bambang et al. 1995; Soegianto et al. 1999a; 1999b; Wu & Chen 2004).

Gresik is one of the big industrial cities in Indonesia. Gresik coastal waters in Indonesia is believed to receive wastewater discharges from a number of wastewater treatment facilities of industries located along the Gresik coastal zone. This zone has been under constant urban pressure during the last fifteen years due to industrial development. The potential industries which contribute to the level of metal in these waters include a superphosphate plant, an asphalt plant, a coal-fired electric power plant, a natural gas processing plant, metal smelters and refineries. These waters also represent the habitat of some edible organisms caught by local fishermen. Many people still use these waters as their fishing ground.

The purpose of this study was to report the concentration of metals in the tissues of aquatic organisms from the Gresik coastal waters and to discuss the implications of such contamination in terms of public health.

Materials and Methods

Five species of fish, i.e. ponyfish (*Leiognathus equlus*), anchovy (*Coilia dusumieri*), milk fish (*Chanos chanos*), 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 local fishermen using gillnets in Gresik coastal waters in June 2003 and June 2004 (Fig. 1).

Samples of fish species whose entire body is consumed such as anchovy and ponyfish were homogenized, and prepared for metal detection. For other fishes and shrimp, only the edible part or flesh was used for metal detection. Before filleting, the external water of each individual sample was absorbed using tissue paper. The flesh was then pooled, 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 also underwent the same process as well as the other species.

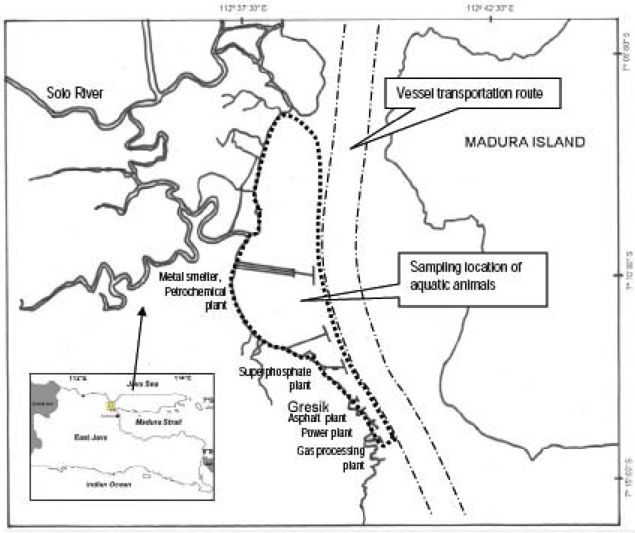

Figure 1. Sampling location of aquatic animals

Each pooled sample was then divided in two to six sub samples, depending on the quantity. The sub-samples were put into special flasks separately, weighed, then frozen at –20°C for a period of not less than 8 hours. The frozen sample was then placed under vacuum on a freeze-dryer unit (Labconco) until the sample was completely dried. Dried sample was weighed and approximately 1 g of each was digested in 20 ml of HNO_3 (ultrapur, Merck) using Microwave Digester (Ethos D) for approximately 25 minutes. Digests were filtered through Whatman no. 4 paper and made up to 50 ml with double de-ionized water. Metal concentrations in the digests are determined by inductively-coupled plasma emission spectroscopy (Thermo Jarrell Ash Type: IRIS Advantage). The metals analyzed were arsenic (As), cadmium (Cd), copper (Cu), zinc (Zn), lead (Pb), chromium (Cr) and nickel (Ni). The accuracy of analytical procedure was monitored by analyses of dogfish muscle reference materials, DORM-2, certified for metals by the National Research Council of Canada. The standard reference material digests were found to conform (95 - 112 %) with the documented values for certified trace metal concentrations.

Results

Composite tissue samples of edible flesh and whole body of organisms were processed and analyzed for various heavy metals. These included five species of fish and one species of shrimp obtained with gill net of local fishermen. Dry weight trace metal concentrations recorded from different species are summarized in tables 1 and 2. We noted that all animals caught in 2003 and 2004 contained Cu and Zn levels higher than the level of other metals (As, Cd, Cr, Ni and Pb) in the same animals. Our findings also showed that the concentration of certain metals (Zn, Cr and Cd) in whole body of animals collected in June 2003 were relatively higher than those recorded in the muscle tissues. In June 2004, we noted that concentrations of Cd, Cr, Cu, Ni and Zn in whole body samples were higher than those in flesh samples.

Table 1. Metal concentrations measured in tissue samples (mg•kg^{-1} dry weight) of animals collected from the Gresik coastal waters in June 2003

Metal	Species					
	Banana shrimp	Ponyfish	Anchovy	Milkfish	Mullet	Sea Catfish
	Penaeus merguensis	*Leiognathus equulus*	*Coilia dusumieri*	*Chanos chanos*	*Mugil vaigiensisi*	*Arius leptonota-canthus*
	Flesh	Whole body	Whole body	Flesh	Flesh	Flesh
	N = 17	N = 14	N = 15	N = 4	N =5	N = 2
	TW = 178.6	TW = 85.8	TW = 100.2	TW = 86.9	TW = 55.6	TW = 38.8
Arsenic	2.23 ± 0.39	1.33 ± 0.11	1.63 ± 0.21	2.50 ± 0.34	1.62 ± 0.01	1.48 ± 0.19
Cadmium	0.08 ± 0.01	0.07 ± 0.02	0.04 ± 0.01	< 0.004	< 0.004	< 0.004
Chromium	< 0.16	0.32 ± 0.08	0.91 ± 0.16	0.22 ± 0.06	0.20 ± 0.02	< 0.16
Copper	26.69 ± 1.41	8.10 ± 0.51	6.78 ± 0.26	8.13 ± 0.60	5.20 ± 0.58	8.82 ± 0.21
Nickel	0.34 ± 0.12	0.33 ± 0.04	0.47 ± 0.08	0.34 ± 0.01	0.36 ± 0.20	0.06 ± 0.00
Lead	0.29 ± 0.08	0.45 ± 0.06	< 0.001	0.21 ± 0.05	2.20 ± 0.40	2.57 ± 0.96
Zinc	52.32 ± 3.13	85.22 ± 0.01	87.15 ± 0.43	17.90 ± 0.45	13.91 ± 1.03	20. 46 ± 1.13
Selenium	0.95 ± 0.37	2.21 ± 0.30	1.78 ± 0.02	1.30 ± 0.23	1.49 ± 0.15	0.54 ± 0.11

Legend: N = number of individuals; TW = total weight (g); < = below detection limit

Table 2. Metal concentrations measured in tissue samples (mg•kg^{-1} dry weight) of animals collected from the Gresik coastal waters in June 2004

Metal	Species					
	Banana shrimp	Ponyfish	Anchovy	Milkfish	Mullet	Sea Catfish
	Penaeus merguensis	*Leiognathus equulus*	*Coilia dusumieri*	*Chanos chanos*	*Mugil vaigiensis*	*Arius leptonota-canthus*
	Flesh	Whole Body	Whole Body	Flesh	Flesh	Flesh
	N = 25	N = 27	N = 18	N = 5	N = 3	N = 4
	TW = 218.4	TW = 192.5	TW = 142.3	TW = 159.2	TW = 102.2	TW = 150.8
Arsenic	1.50 ± 0.42	1.03 ± 0.02	0.50 ± 0.09	0.62 ± 0.08	0.005 ± 0.003	1.67 ± 0.21
Cadmium	0.03 ± 0.02	0.009 ± 0.003	< 0.004	< 0.004	< 0.004	< 0.004
Chromium	0.21 ± 0.03	0.79 ± 0.12	0.83 ± 0.12	< 0.16	0.22 ± 0.06	< 0.16
Copper	24.49 ± 5.30	7.85 ± 0.22	6.97 ± 0.89	3.11 ± 0.76	2.15 ± 0.86	2.34 ± 0.23
Nickel	0.11 ± 0.04	0.51 ± 0.07	0.20 ± 0.01	0.13 ± 0.04	0.004 ± 0.003	0.003 ± 0.002
Lead	0.31 ± 0.02	0.40 ± 0.11	0.25 ± 0.14	0.29 ± 0.04	0.09 ± 0.03	0.20 ± 0.09
Zinc	47.25 ± 4.95	25.73 ± 1.75	77.19 ± 4.22	11. 25 ± 1.02	2.09 ± 0.16	21.21 ± 2.41
Selenium	0.25 ± 0.11	0.13 ± 0.03	< 0.001	0.54 ± 0.07	0.02 ± 0.01	0.37 ± 0.03

Legend: N = number of individual; TW = total weight (g); < = below detection limit

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 concentrations of copper and zinc in both fish and shrimp collected in 2003 and 2004 were relatively higher compared to concentrations of other metals (As, Cd, Cr, Ni and Pb) in the same animals. The similar findings were also recorded in crustaceans (Ridout et al. 1985; Swaileh & Adelung 1995; Parsons 1998; Hossain & Khan 2001; Miramand et al. 2001; Tyrrell et al. 2005) and fish (Parsons 1999; Miramand et al. 2001; Zehra et al. 2003; Tyrrell et al. 2005) caught from other waters of the world. The copper level recorded in the flesh of shrimp samples (*Penaeus merguensis*) showed elevated copper concentration than that in other fish. 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 & Rainbow 1982; Rainbow 1995; Soegianto et al. 1999a).

Almost all metals measured in this study have relatively lower values than the values recorded in aquatic animals from other regions of the world. Only the level of zinc was comparable to the results of other researchers. The findings of other studies are summarized in table 3, and are compared with the concentrations reported in this study and elsewhere in the world.

As reported in this study, trace element concentrations varied markedly among species. These variations are presumably due to individual sampled 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 had higher concentrations than more pelagic species, which may be related to greater exposure to contaminated sediments (Trucco et al. 1990; Parsons 1999).

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 concentration of certain metals (Cd, Cr and Zn) in whole body of fish (anchovies and ponyfish) collected in June 2003

Table 3. A comparison of metal concentrations (mg•kg^{-1} dry weight) in aquatic animals recorded from the 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)	1.08-2.62	0.01-0.09	<0.16-0.24	19.19-29.79	0.07-0.46	0.21-0.37	0.14-1.32	42.30-55.45	Present study
	Leiognathus equulus (Teleostei)	1.01-1.44	0.006-0.09	0.24-0.91	7.63-8.61	0.29-0.58	0.39-0.51	0.10-2.51	23.98-85.23	Present study
	Coilia dusumieri (Teleostei)	0.41-1.84	<0.004-0.05	0.71-1.07	6.52-7.86	0.19-0.55	<0.001-0.39	<0.001-1.80	72.97-87.58	Present study
	Chanos chanos (Teleostei)	0.54-2.84	<0.004	<0.16-0.28	2.35-8.73	0.09-0.35	0.16-0.33	0.47-1.53	10.23-18.35	Present study
	Mugil vaigiensis (Teleostei)	0.002-1.63	<0.004	0.18-0.28	1.29-5.78	0.001-0.56	0.06-2.60	0.01-1.64	1.93-14.94	Present study
	Arius leptonotacanthus (Teleostei)	1.29-1.88	<0.004	<0.16	2.11-9.03	0.001-0.06	0.11-3.53	0.34-0.65	19.33-23.62	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 belengerii (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 & 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 & 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 & Adelung (1995)

Table 3. A comparison of metal concentrations (mg•kg^{-1} dry weight) in aquatic animals recorded from the Gresik coastal waters and other regions of the world (continued)

Location	Species	As	Cd	Cr	Cu	Ni	Pb	Se	Zn	References
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)

were relatively higher than those documented in muscle tissue of mullet, milkfish and sea catfish. In 2004, we noted that concentrations of Cd, Cr, Cu, Ni and Zn in whole body samples were higher than those in flesh samples.

Various countries and organizations have released standards for maximum limits of metals in the muscle tissue of marine biota. By using those standards, we discuss the implications of such contamination in terms of public health. According to the standard presented in table 4, almost all animals collected from the Gresik coastal waters contained trace metals in their tissue within acceptable range for consumption.

Table 4. Maximum residue limit and maximum permitted concentration of metals in marine biota muscle (mg•kg^{-1}) from various countries and organizations

Metal	U.K.[1]		Australia [2]		Hong Kong [1]		IRPTC [3]		European Regulation 466/2001/ EC [4]		Indonesia [5]	
	ww	dw	ww	dw	ww	dw	ww	dw	ww	dw	ww	dw
Cadmium	-	-	0.20	0.80	2	8	-	-	0.1	0.4	-	-
Lead	-	-	1.5	6.0	6	24	-	-	0.4	1.6	2	8
Selenium	-	-	1	4	-	-	-	-	-	-	-	-
Chromium	-	-	-	-	1	4	-	-	-	-	-	-
Copper	20	80	-	-	-	-	-	-	-	-	20	80
Zinc	50	200	-	-	-	-	-	-	-	-	100	400
Arsenic	-	-	1+	4+	-	-	-	-	-	-	1	4
Nickel	-	-	-	-	-	-	0.5	2	-	-	-	-

Legend: ww = wet weight; dw = dry weight; Wet weights (ww) were transformed to dry weight (dw) by multiplying by a factor 4 (Parsons 1998; 1999); +Inorganic arsenic, this study measured total arsenic. [1]Parsons (1998; 1999); [2]Otway (1992); [3]IRPTC (1988); [4]Tyrrell et al. (2005); [5]Decree of General Director of Food and Drug Supervision No. 03725/B/SK/VII/89 concerning maximum limit of metals in food

Conclusion

Gresik coastal waters in Indonesia receive wastewater discharges that may contain metals from a number of industries located along the Gresik coastal zone. However, the concentration of metals in aquatic animals collected from this area was within the acceptable range for consumption. It is recommended that constant monitoring should be conducted to avoid the intake of contaminated aquatic animals.

Acknowledgements

This study was supported by the Research Institute of Airlangga University. Special thanks to Mrs. Mochammad Affandi, Miftakhul Ulum and Eko W. Putro for helping with sample collection. The authors also thank the anonymous referees for their valuable comments on the manuscript.

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Received: 26 May 2006; Accepted: 25 January 2008