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Heavy metal contamination in Indonesian coastal marine ecosystems: A historical perspective

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Abstract—Toxic heavy metals are one of the widespread environmental contaminants in Indonesian coastal waters. Research and monitoring on heavy metal contaminations have been conducted since 1980. The development of pollution studies may be divided into three phases: firstly, monitoring of heavy metal contamination focused on seawater (1979–1990); secondly, research and monitoring focused on biota and sediment (1990–2000); and the third phase, the research focused on bioassay and geochemistry of heavy metals (2000–present). Most metals have been intensively studied in components of coastal ecosystem. An elevated heavy metal contamination has been mostly recorded in the northern coast of Java Island and the eastern coast of Sumatra Island, while coastal waters of Borneo and the Sulawesi Islands are, in general, relatively pristine. The concentration of heavy metals in water and biota is commonly very low. In contrast, heavy metals in sediments are relatively elevated in concentrations especially in areas where land-based activities exist such as industrial activities. Recent research has been directed on metal speciation in sediments and development of bioassay using local species of organisms to reduce uncertainty when local managers have to make a decision on environmental issues related to ecological risk assessment.

Key words: heavy metals, coastal waters, sediments, biota, Indonesia

Introduction

Population growth and industrial development along the coast have greatly influenced pollution loads into coastal marine environment. Land-based activities are major source of pollution in the coastal waters of Indonesia. Based on several studies conducted in coastal areas, heavy metal contaminations are among the persistent issues in Indonesia. These heavy metals have potential negative impact on the health of marine living resources and the people who consume seafood products.

Fate of metal contaminants in coastal ecosystem can be in the three compartments, seawater, sediments and biota (Fig. 1), and the development of research on this subject followed ideas that were developed during the last three decades. Metal pollution studies in water and sediments have been conducted by many researchers since early 1980s. Studies of metal pollution based on concentration of metals in solute form can be really challenging especially when availability of instruments and laboratory that meet the state of the art technology is limited. Because metal concentration in seawater is highly fluctuated, it is difficult to make a general trend of metal contaminants in a wide area. Hence, research of metal pollution has been directed to study of metals in sediments since 1990s. The first review of toxic trace metal pollution in sediments provided information on trace metal contamination in several coastal sediments of Indonesia (Arifin 2001).

Solid and strategic actions are urgently needed to mitigate the ever increasing metal contaminants in both biota and

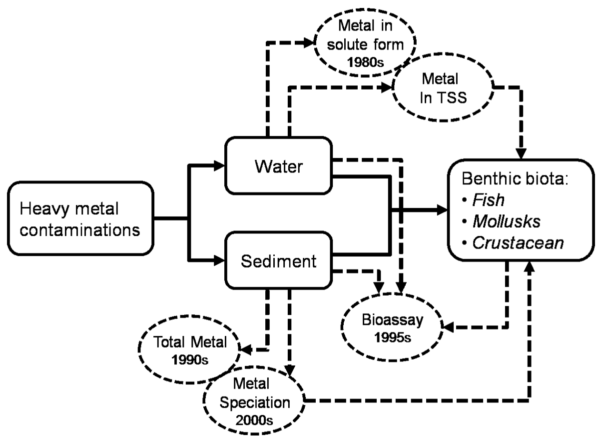

Fig. 1. Fate of metal contaminants in coastal ecosystem (solid arrow) and the development of research during the last three decades (broken arrow).

sediments. In this review paper, we try to provide historical perspective of heavy metal contamination and eco-toxicological studies that have been done in Indonesia, and efforts in reducing coastal pollution.

Metal contaminants in seawater

Heavy metals in seawater can be divided into two major components, i.e., metals in solute form and metals that bind to suspended particles or sediments (phytoplankton, zooplankton, debris, clay and silts). Toxic trace metals in coastal waters, especially Hg, Pb and Cd have been measured in some components of coastal waters (Sanusi unpubl., Mulyanto unpubl.), but knowledge on other trace metals such as As, Cr and Sn is very limited. Research of toxic metals in water was mainly represented by measuring metals in solute form which was commonly found to be very low in concentration.

The concentrations of metals in solute form are mostly lower than that of national water quality standard (Table 1). These low concentrations of heavy metals in solute form can be due to high metal sorption by suspended particulate matter, phytoplankton and mollusks or seaweed cultures. For example, an increasing primary productivity and intensive green mussel's culture along the coast of Jakarta indicated that heavy metals in solute form were relatively low in concentrations (Arifin 2008a). Only in certain cases the concentrations of toxic trace metals such as Cd in waters was above threshold concentration of the national seawater quality standard in Jakarta Bay (Arifin and Fitriati 2006).

Considering the availability of instruments, monitoring activities have focused on both in the seafood products and on the sediment of coastal system. Monitoring of trace metals in biota is directly used to predict risk of contaminants to human and to study possible biomagnification processes. On the other hand, research on heavy metals in sediments has implications to understand various possible effects of metal contaminants on biota those are exposed to their habitat conditions.

Metal contaminants in sediments

Sediments can be used as an indicator of pollution due to its role as a sink of various contaminants. Measurement of heavy metals in sediments provided better indicator than those in solute form (in water), and the concentration of heavy metals in sediments was relatively stable (Table 2). In contrast to the toxic trace metals in solute form, the concentrations of metal in sediments showed an increasing trend during the period of 1985–2000 and concentrations of trace metals such as Pb and Cd were relatively higher compared to background levels, especially in coastal areas bordering the

Table 1. Metal in solute form (ppm) in several coastal waters of Indonesia.

Coastal waters	No. St	Sampling time	Pb	Cd	Cu	Ni	Zn	Ref
Coastal waters around Fishery harbour of Pelabuhan Ratu	8	2002, May	<0.001–0.003	<0.001	<0.001–0.002	<0.001–0.001	<0.001–0.003	Anindita (2002)
Estuary of Digul river and Arafura sea	26	2001, May	<0.001–0.015	0.001–0.002	<0.001			Hutagalungand Manik. (2002)
Jakarta Bay	23	2003, July	0.001–0.003 (0.0005)	<0.001	0.001–0.002 (0.0004)	0.001–0.002 (0.0002)	0.001–0.030 (0.0033)	Arifin et al. (2003)
Coastal waters of Banten Province	23	2001, April	<0.001–0.018 (0.006)	<0.001–0.001	<0.001–0.001 (0.001)	<0.001–0.004 (0.001)	<0.001–0.018 (0.005)	Rochyatun et al. (2005)
Jakarta Bay	23	2004, Jan.	0.001–0.005 (0.002)	<0.001	0.001–0.002 (0.001)	0.001–0.004 (0.003)	0.001–0.017 (0.006)	Unpublished data
Coastal waters of Cirebon City (North Java)	7	2004, Nov.	0.003–0.004		0.001–0.002		0.006–0.016	Hidayat (2005) in Nugroho (2010)
Kampar River, Riau	3	2006, July, Sept.	0.011–0.017	0.035–0.046				Erlangga (2007)
Delta Berau, East Kalimantan	17	2007, April	nd–0.002		nd–0.002			Arifin (2008)

nd: not detected or concentration <0.001 ppm.

Table 2. Concentrations range and average of several heavy metals in sediments (mg kg^{-1} dw) of several coastal waters of Indonesia.

Coastal waters	No. St	Sampling time	Hg	Pb	Cd	Cu	Zn	Ref
Coast of Pantai Ujung Watu (Central Java)	11	Sep 1983, Jan and Nov 1984	0.01–0.21	1.1–10.9	0.11–0.41	2.1–29.6	0.6–9.3	Razak (1986)
Estuary of Brantas river and Solo river	12	Jul–Aug Nov–Dec 1984		9.0–23.0	0.06–0.42	24.0–58.0	40.0–122.0	Everaarts (1989)
Jakarta Bay	13	Jun, Nov–1990	0.13–1.63 (0.55±0.43)	79.5–176.5 (101.3±26.0)	0.90–2.66 (1.74±0.57)	7.2–53.8 (27.5±13.45)		Hutagalung (1994)
Estuary of Way Kambas dan Way Sekampung	7	Jul–1998		7.4–12.6	nd–0.01	1.6–6.1	17.5 - 32.9	unpublished data
	7	Sep–1998		10.7–16.1		1.9–6.0	18.5–39.8	
Estuary of Digul dan Arafura Sea	26	May–2001		3.6–12.4 (7.8±2.14)	<0.1			Hutagalung and Manik (2002)
Telaga Tujuh Coast (Kep Riau)	5	July 2002		82.5–98.3 (88.2)		23.7–71.6 (46.3)	48.2–149.3 (96.8)	Amin (2002)
Pelabuhan Perikanan (Pel. Ratu, West Java)	8	May–2002		7.7–28.9	0.07–0.34	12.9–47.4	72.1–145.4	Anindita (2002)
Jakarta Bay	23	Jul–2003		5.7–42.9 (22.51)	0.04–0.50 (0.18)	8.6–186.8 (46.1)	51.9–480.5 (172.80)	Arifin et al. (2003)
	23	Jan–2004		3.2–57.8 (18.7)	0.01–0.28 (0.11)	4.8–76.8 (24.1)	4.8–408.5 (139.5)	unpublished data
Berau Delta	21	April 2008		3.9–18.7 (12.2±4.63)	0.02–0.12 (0.06±0.02)	1.58–34.1 (16.5±9.38)	1.1–9.0 (6.1±2.17)	Arifin (2008b)

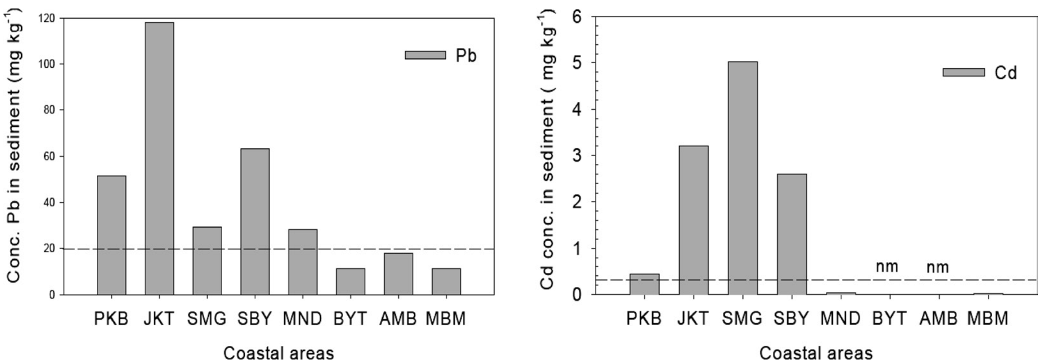

Fig. 2. Lead (Pb) and Cadmium (Cd) concentrations (mg kg^{-1}) in sediment coastal cities during period 1985–2000 (Arifin 2001). PKB: Pekan Baru, JKT: Jakarta, SMG: Semarang, SBY: Surabaya, MND: Manado, BYT: Buyat, AMB: Ambon, and MBM: Membramo, nm: not measured. ---: average metal in shale.

provincial cities in northern coast of Java Island, namely Jakarta, Semarang and Surabaya (Fig. 2). Increasing industrial areas, harbor developments and population growth contribute to the increasing trend of metal contaminants during early 1980s to 1990s (Arifin 2001, 2004).

Highly toxic trace metals in sediments indicated that metals in water column might mostly be bound to particulate matter and settled not far from the shore. Once contaminants

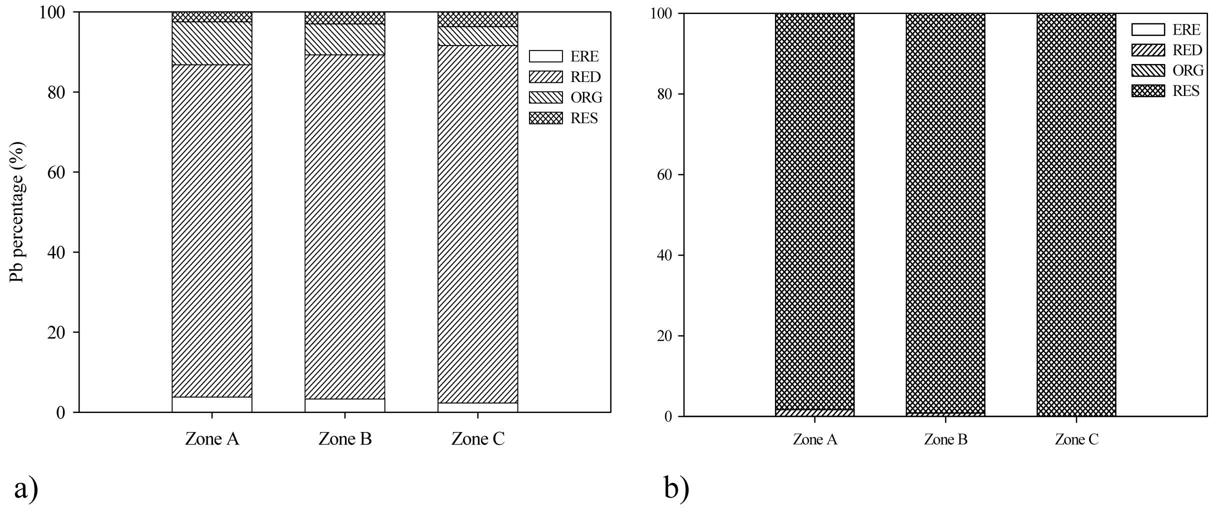

Fig. 3. Lead (Pb) speciation in sediments of (a) Jakarta Bay and (b) Berau Estuary (Arifin et al. 2010). Metal speciation in sediment occured in four fractions, i.e., ERE: easily reducible, RED: redicuble, ORG: organic and RES: residual.

settled in sediments, they are potentially absorbed by benthic organisms that can transfer the metals from sediments to higher trophic levels of food chain (Rees et al. 1999). In the long run, this will become potential problems to the quality of pelagic system. Recent studies showed that metal concentrations in sediments do not fully provide information on metal bioavailability. Metal bioavailability to benthic organnisms depends on various factors, such as geochemical history of sediments (Wang et al. 2002), animal behavior (Bendell-Young and Arifin, 2004) and exposure duration. Early works on metal speciation in Indonesian coastal waters were done by Takarina et al. (2004) in Semarang harbor, followed by Amin et al. (2009) in Dumai coastal waters; Takarina (2010) in Jakarta Bay; Nugroho et al. (2010) and Arifin et al. (2010) in Berau Delta.

Heavy metal speciation showed different patterns under different coastal ecosystems. In contaminated sediments of Jakarta Bay, Pb fraction is generally associated with reducible fraction (Fig. 3a), while in uncontaminated sediments of Berau Delta most Pb was in residual fraction which was not potentially available to benthic biota (Fig. 3b).

Metal contaminants in biota

Early study on heavy metal concentrations in marine biota was done in Angke Estuary in North Jakarta in 1979 and 1980 on various seafood resources such as fish, shrimp *(Penaeus monodon)* and mollusks (*Anadara granosa*; *Perna viridis*), in which concentrations of the heavy metals were mostly below WHO's threshold concentrations (Hutagalung and Razak 1982, Hutagalung 1987, Hutagalung et al. 1989). Recently, studies on butyltin in marine biota (Sudaryanto et al. 2000, 2002, Rumengan et al. 2008), total Hg in the Bay of Ratatotok (Limbong et al. 2003, 2004) and in Buyat Bay

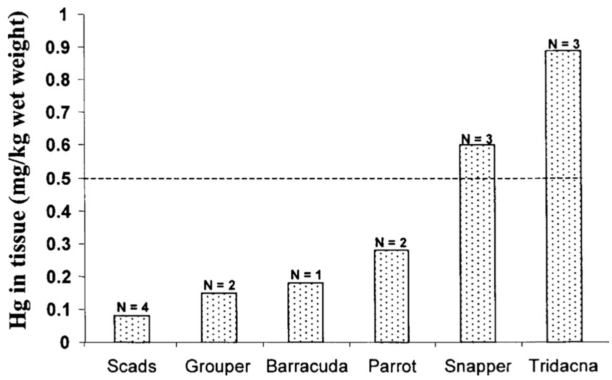

Fig. 4. Mercury concentrations in various fish from Ratatotok Bay where traditional mining situated at the upland area (Limbong et al. 2004).

(Lasut et al., 2010) were also done. The Hg concentrations in fishes were mostly below the maximum recommended concentrations ($0.5\,mg\,kg^{-1}$ ww), except those in the snapper and giant clam (*Tridacna*) which showed the Hg levels above the limit of maximum concentration (Fig. 4).

The concentrations of Pb and Cd in mussel tissues (*Perna viridis*) were generally higher in small- or non-marketed size mussels ($<$4 cm shell length) than in those of large- or commercial size ($>$5 cm SL). Moreover, the concentration of heavy metals in mollusks was generally 2–3 times higher than that in fishes (Arifin 2008a, b). These general trends would suggest that marine biota at small sizes are physiologically more active than the large size groups with respect to the accumulation of the particular heavy metals. Furthermore, feeding behavior and habitat preferences of biota may contribute to the accumulation of metals.

Table 3. Several contaminants toxicity tests that were done on local species during the period of 1994–2008.

Test species	Test material	Life stage	Test duration	Test System	Effect measured	Conc.	References
Plankton							
Chaetoceros gracilis	F	—	96-h	S	IC50	$>$10000 mg L^{-1}	Darmayati et al. 1999
C. gracilis	Cd		96-h	S	IC50	2.61 mg L^{-1}	Hindarti et al. 2008
C. gracilis	Cd	—	96-h	S	IC50	1.5 mg L^{-1}	Puspitasari and Hindarti 2009
C. gracilis	Cd		96-h	S	IC50	1.9 mg L^{-1}	Hindarti et al. 1999
Tetraselmis sp	Cd	—	96-h	S	IC50	5.79 mg L^{-1}	Hindarti et al. 1999
Tetraselmis sp	F		96-h	S	IC50	24.18 mg L^{-1}	Darmayati et al. 1999
Tetraselmis sp	Effluent glass industry	—	96-h	S	EC50	56.5%	Darmayati et al. 1999
Fish							
Chanos chanos	Cr (VI)	Larvae	7 days	S, R	LC50	25.6 mg L^{-1}	Panggabean et al. 1999
C. chanos	Cd	Larvae	96-h	S	LC50	5.9 mg L^{-1}	Panggabean et al. 1999
Chromis viridis	Cyanides	Juvenile	96-h	S, R	LC50	41.3 μg L^{-1}	Arifin and Hindarti 2006
C. viridis	Cd	Juvenile	96-h	S, R	LC50	13,3 mg L^{-1}	Arifin and Hindarti 2006
Cromileptes altivelis	Cd	Juvenile	96-h	S	LC50	2.16 mg L^{-1}	unpublished data 2008
Musels and Sea urchin							
Perna viridis	Cr (VI)	Larvae	24-h	S	EC50	4.5 mg L^{-1}	Panggabean 1997
P. viridis	Effluent glass industry	Larvae	24-h	S	EC50	13.6%	Darmayati et al. 1999
P. viridis	Cd	Larvae	24-h	S	EC50	1.2 mg L^{-1}	Panggabean, 1997
P. viridis	F	Larvae	24-h	S	EC50	7.7 mg L^{-1}	Darmayati et al. 1999
Tripneustes gratilla	Cd	Larvae	20 min	S	EC50	1.65 mg L^{-1}	Hindarti et al. 2008

Note: F: Flouride, S: static, R: renewal, EC: exposure concentration, LC: lethal concentration, IC: inhibition concentration.

Bioassay for seawater and contaminated sediments

Bioassay and toxicity studies using local biota were developed during the ASEAN-Canada Marine Science Program (1993–1998). During this time, many local species, such as those of phytoplankton, sea-urchin eggs, shrimp larvae, green mussel larvae, milkfish fry, and grouper fry were used to develop the techniques for toxicity assessment (Table 3). At present, these biota have been well established as test organisms. However, the progress on the development and refinement of toxicity studies after the termination of ASEAN-Canada Marine Science Program was relatively stagnant. In recent years, a few laboratories are carrying out routine assessment of toxicities and development of marine contaminated sediment's bioassay, such as Ecophysiology Lab at Faculty of Fisheries and Marine Science–IPB (Dr. Etty Riani, pers. comm.) and Ecotoxicology Lab at Research Centre for Oceanography–LIPI. Our laboratory used two species that are being developed as test organisms (amphipod and Java Medaka).

Future direction of contaminant research

The marine environmental issues that Indonesia currently faces are summarized as follows: (a) unbalanced population distribution within the country has put more pressure to the coastal and marine ecosystems leading to an increase in risk of contaminants exposure to human health; (b) unsustainable practices of extraction of non-renewable resources have caused many conflicts between the interest of preserving environment and economic development; (c) oil and gas industries and mining industries have been rising in the last 10 years, and (d) high biodiversity of marine and coastal waters are in threat but coastal communities heavily depend on it. To address these issues, the research should focus on the following three areas, possibly integrating with biodiversity issues: (1) fate and effect of contaminants (POPs and heavy metals) on both at species and community levels, (2) geochemistry of heavy metals and selection of tropical species for standard bioassays, and (3) bioremediation of the contaminants in coastal systems. These areas of research are a central issue in understanding contaminants behavior in nature.

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