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Seasonal distribution and geochemical fractionation of heavy metals from surface sediment in a tropical estuary of Jeneberang River, Indonesia



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ABSTRACT

Lead (Pb) and zinc (Zn) concentrations from surface sediments were determined at 17 stations in riverine, estuarine, and marine environments during the wet and dry seasons. The geochemical fractionations were obtained by a sequential extraction procedure in five geochemical forms: exchangeable, bound to carbonate, iron–manganese oxide, organic, and residual fractions. The concentrations of Pb and Zn in the water were higher during the wet season than the dry season and higher in the riverine and marine samples than the estuarine samples. Following geochemical fractionation, the metals were found mainly in the residual fraction. The results indicated that the concentrations were influenced by season, with the sources of Pb and Zn derived mainly from rivers and natural sources. The contamination status in the estuary of the Jeneberang River was classified as weak to moderate pollution and the risk level to aquatic biota attributed was no risk to low risk.

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Heavy metals are derived from natural and anthropogenic sources. In most cases, heavy metals of anthropogenic origin are released from urban and industrial areas adjacent to rivers and estuaries. Heavy metal contamination in the coastal marine environment has become a serious problem in Asian coastal megacities as they undergo rapid economic growth and industrialization (Jiang et al., 2015). Makassar, as the largest city in eastern Indonesia, is undergoing rapid economic growth and industrialization. The Jeneberang River flows through Makassar City, including the upstream area in the Bawakaraeng Mountains, Gowa district and the downstream area in the Makassar Strait. The Jeneberang River flows through village and city regions and therefore has the potential for contamination by various metals from agricultural, domestic, and transportation sources. There is also potential intake of metals from marine sources such as the Makassar international seaport, ship docking and manufacturing, hospital, and industrial sectors (Samawi, 2007; Taufiq, 2010; Werorilangi et al., 2013).

Heavy metals in river water are a concern due to their persistence in the environment, bioaccumulation, and high levels of toxicity (Hu et al., 2013; Wang et al., 2015). Assessment of heavy metal concentrations in sediment are useful to determine pollution levels in estuarine and marine environments and to provide basic information for determination of environmental health risks (Hu et al., 2013). In sediments, heavy

metals can exist in various chemical forms, which exhibit different physical and chemical behaviors with respect to their chemical interactions, mobility, biological availability, and potential toxicity (He et al., 2009). Heavy metals interact with sediment matrices and exhibit different binding mechanisms with carbonates, iron (Fe) – manganese (Mn) oxides, organic matter, and the lattices of refractory crystalline minerals such as silicates (Gao and Li, 2012). Determination of the geochemical fractionation of heavy metals in sediments is important to assess their potential toxicity, threat to the ecosystem, and source (natural processes or anthropogenic sources). The accumulation of heavy metals in the resistant fraction is an indication that they originate from natural processes, while accumulation in the non-resistant fraction indicates they originate from anthropogenic sources (Liu et al., 2015).

Several studies on the distribution and geochemical fractionation of heavy metals in sediment have been published (Armid et al., 2014; Takarina et al., 2004); however, limited information is available due to seasonal effects. The concentrations of heavy metals during all four seasons have been reported in the Gulf of Aqaba, Jordan (Al-Najjar et al., 2008), southeast coast of the Caspian Sea (Bastami et al., 2015), and Yellow Sea (Jiang et al., 2015); however, these locations are not tropical.

A seasonal study is needed, because the seasons differentially influence the physico-chemical parameters within the environment. Changes in environmental conditions, such as pH, temperature, and redox potential (Eh), could influence metal release from the solid to liquid phases that transformed sediments from the metal main sink to the sources for the overlying waters (Wang et al., 2015).

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The objectives of this study were to investigate the distribution and geochemical fractionation of heavy metals in surface sediments during different seasons (wet and dry) to identify the possible sources of heavy metals and to assess the pollution levels and ecological risks associated with lead (Pb) and zinc (Zn).

A total of 17 surface sediment samples were collected from riverine, estuarine, and marine waters during January and August 2015, which represent the wet and dry seasons, respectively. The riverine zone contained sampling stations R1–R3, the estuarine zone contained sampling stations E1–E7, and the coastal (marine) zone contained sampling stations CS1–CS7. Sample locations were determined by a global positioning system (Fig. 1). The surface sediments were sampled using an Ekman grab sampler. At each sampling site, the top 0–10 cm of sediment were collected using a plastic spatula, and each sample was packed in a new polyethylene zipper bag. After collection, the samples were stored and transported to the laboratory in a cooler box with ice packs and then stored in a fridge at 4 °C prior to analysis. In the laboratory, samples were first sieved through a 2 mm nylon sieve to remove coarse debris and shell fragments, ground using an agate mortar and pestle in a fume cupboard, and then sieved through a 0.5 mm sieve to remove large particles and homogenize the mixture.

The geochemical fraction of heavy metals was determined using sequential extraction procedure (Tessier et al., 1979). This method provides the exchangeable, bound to carbonates, bound to Fe—Mn oxides, bound to organic matter and residual fractions with the following procedures;

- i. Exchangeable (F1). The sediment was extracted at room temperature for 1 h with 8 mL of either magnesium chloride solution (1 M $MgCl_2$, pH 7.0) or sodium acetate solution (1 M NaOAc, pH 8.2) with continuous agitation.
- ii. Bound to carbonates (F2). The residue from (i) was leached at room temperature with 8 mL of 1 M NaOAc adjusted to pH 5.0 with acetic

acid (HOAc). Continuous agitation was maintained and the time necessary for complete extraction was evaluated.

- iii. Bound to Fe—Mn oxides (F3). The residue from (ii) was extracted with 20 mL of either 0.3 M $Na_2S_2O_4$ + 0.175 M Na-citrate + 0.025 M H-citrate or 0.04 M $NH_2OH-HCl$ in 25% (v/v) HOAc. The latter experiments were performed at 96 ± 3 °C with occasional agitation and the time needed for complete dissolution of the free iron oxides was evaluated.
- iv. Bound to organic matter (F4). The residues from (iii) were added 3 mL of 0.02 M HNO_3 and 5 mL of 30% H_2O_2 adjusted to pH 2 with HNO_3 , and the mixture was heated to 85 ± 2 °C for 2 h with occasional agitation. A second 3-mL aliquot of 30% H_2O_2 (pH 2 with HNO_3) was then added and the sample was heated again to 85 ± 2 °C for 3 h with intermittent agitation. After cooling, 5 mL of 3.2 M NH_4OAc in 20% (v/v) HNO_3 was added and the sample was diluted to 20 mL and agitated continuously for 30 min. The addition of NH_4OAc is designed to prevent adsorption of extracted metals onto the oxidized sediment.
- v. Residual (F5). The residue from (iv) was digested with a HF— $HClO_4$ mixture according to the procedure described below for total metal analysis.

The “selective” extractions were conducted in centrifuge tubes (polypropylene, 50 mL) to minimize losses of solid material. Between each successive extraction, separation was effected by centrifuging at 10,000 rpm (12,000 g) for 30 min. The supernatant was removed with a pipet and analyzed for trace metals, whereas the residue was washed with 8 mL of deionized water; after centrifugation for 30 min, this second supernatant was discarded. The volume of rinse water used was kept to a minimum to avoid excessive solubilization of solid material, particularly organic matter. Deionized water used in preparing stock solutions and in each step of the leaching procedure was obtained from a

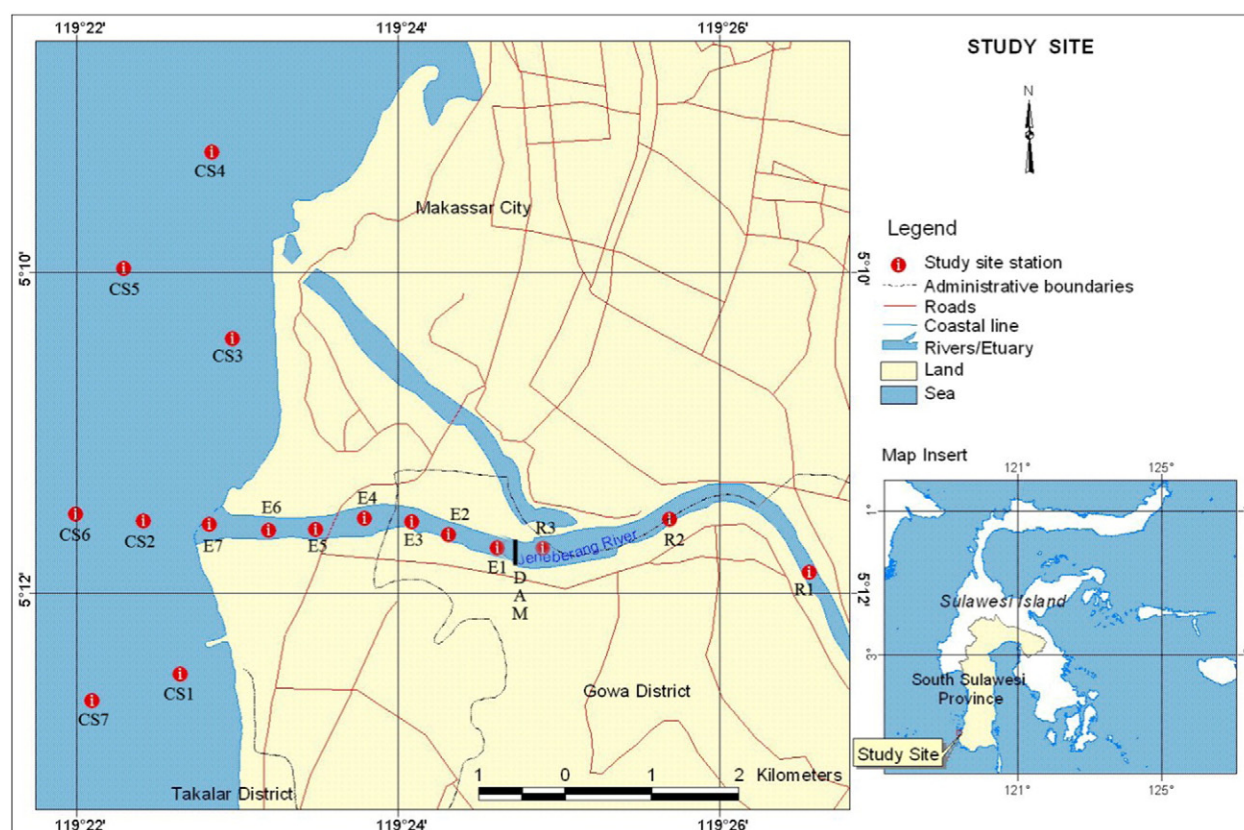


Fig. 1. Location of sample sites.

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