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Assessment of Metal Pollution Status Through the Fractionation and Bioavailability of Lead Metal (Pb), Chromium (Cr), and Mercury (Hg) in Sediments in the Islands Water Areas of North Maluku Province, Indonesia

Inayah[1], Muhammad Farid Samawi[2]*, Khusnul Yaqin[3], Najamuddin[4]

[1]Faculty of Fisheries and Marine Science, Khairun University, Ternate
[2]Department of Marine Science, Faculty of Marine Science and Fisheries, Hasanuddin University
[3]Aquatic Resources Management Program, Faculty of Marine Science and Fisheries, Hasanuddin University
[4]Faculty of Fisheries and Marine Science, Khairun University, Ternate

***Corresponding Author: faridsamawi@unhas.ac.id**

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ABSTRACT

This study was conducted in the waters of North Maluku with various anthropogenic activities that can be sources of metals, namely the Domestic Port in Bastiong, the power plant area on Tidore Island, the Oil Port in Jambula and the Gold Mine in Kao Bay. This study aimed to assess the status of metal pollution through the fractionation of Pb, Cr and Hg metals in sediments related to the bioavailability of Pb, Cr and Hg metals in *Anadara granosa*. Sediment metal fractionation was determined by the Bureau Commune de Reference Sequential Extraction method and metal bioavailability using *Anadara granosa* shells. The results of measuring the total concentration of Pb, Cr and Hg metals in North Maluku waters sediments ranged from 7.98 - 15.11mg/ kg; 0.013-0.317mg/ kg and 0.003-0.0192mg/ kg with Pb, Cr and Hg metal content in *Anadara granosa* of 5.52-10.68mg/ kg; 0.204-0.292mg/ kg and 0.0002-0.0003mg/ kg. Available fractions (F1, F2 and F3) were found in Pb and Cu metal types at domestic ports, power plant ports and oil ports. The residual fraction (F4) of Pb, Cr and Hg dominated at all locations, mostly found in sediments around gold mining. The bioavailability of metals was found to be low, indicating that the source of Pb, Cr, and Hg metal pollution in the water sediments of North Maluku Province predominantly comes from natural sources, such as rocks, which have very low availability for biota.

INTRODUCTION

The main anthropogenic activities in the waters of North Maluku Province are mining, harbor and industry which are often found in North Halmahera, Ternate Island and Tidore. These anthropogenic activities have an impact on reducing the quality of the marine environment and pollution. This research was conducted in the waters of North Maluku which consist of various anthropogenic activities that can be a source of metals, namely the Domestic Harbor in Bastiong, the Power Plant area on Tidore Island, the Oil Harbor in Jambula and the Gold Mine in Teluk Kao.

The total metal concentration is commonly utilized to evaluate metal pollution in sediments; however, it does not adequately reflect or provide insights regarding the mobility, bioavailability, and toxicity of metals. Metal geochemical fractionation of metals is critical in identifying whether metal sources are natural or anthropogenic. Additionally, it offers more precise information regarding the availability of metals to marine biota **(Bastami *et al*., 2018)**.

According to **Sarkar *et al.* (2014)**, the BCR method (Bureau Commune de Reference) analyzes metals in four binding phases. These phases include the acid-soluble phase, which has the least stable bonds with sediments and soils; the reducible phase, which binds to manganese and can be easily separated from sediment under changing conditions from oxic to anoxic; the oxidizable phase, which indicates the amount of heavy metal bound to organic materials and sulfides; and the residual phase, which is a non-bioavailable phase.

Bioavailability refers to the proabortion of metal that can be absorbed by biota. According to **Werorilangi (2019)**, bioavailability of metals in water indicates that metals are present as free ions, which can be readily absorbed by organisms. When metals are in sediment, they participate in geochemical fractions, determining their bioavailability to biota. The geochemical fraction in sediment that is bioavailable will dictate the extent of metal absorption by organisms. This bioavailability influences bioaccumulation on the toxicity impacts on organisms, particularly benthic species **(Paller & Knox, 2013; Werorilangi, 2019)**. Fractionation of metals in sediments in acid-soluble (F1), reduced (F2), oxidized (F3) and residual (F4) forms. The bioavailable forms are F1, F2 and F3, while F4 is difficult to utilize. The level of contamination of heavy metals Pb, Cd and Cu in shellfish follows the bioavailability of these heavy metals in sediment **(Samawi *et al*., 2020)**. Bioavailability studies of As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Zn metals in contaminated coastal sediments from the Berre lagoon, France, showed correlations with potentially bioavailable and bioaccumulated concentrations in the polychaete *Alitta succinea*.

High metal concentrations in living organisms can lead to altered sensory perception and reduced responses to normal olfactory cues, manifesting as hypoactivity, hyperactivity, and diminished swimming ability or resistance to currents. Metals can also cause morphological abnormalities, neurophysiological disorders, genetic mutations, teratogenesis, and carcinogenesis. Furthermore, metals can affect enzymatic and hormonal activities, growth rates, and mortality rates **(Bubb *et al*., 1991; Lestari, 2017)**.

This study aimed to describe the status of the waters of North Maluku Province through geochemical fractionation analysis of Pb, Cr and Hg metals in sediments and the bioavailability of Pb, Cr and Hg metals using the organism *Anadara granosa*.

MATERIALS AND METHODS

This research was conducted in the waters of North Maluku, covering four stations, namely Bastiong Domestic Harbor, Power plant area in Tidore Island, Jambula Oil Harbor, and Gold Mining in Kao Gulf. Sediment and *Anadara granosa* sampling was carried out at the same station with three replications. Measurements of environmental parameters of temperature, salinity, dissolved oxyen (DO), pH, and Eh were carried out *in situ*. Preparation of sediment and *Anadara granosa* samples was carried out at the Chemistry Oceanography Laboratory of the Faculty of Marine Sciences and Fisheries, Hasanuddin University. Meanwhile, metal concentration measurements were carried out at the Testing Laboratory of the Plantation Products, Metal Minerals, and Marine Industry Standardization and Service Agency, Makassar City. The research location and sampling stations are presented in Fig. (1).

Fig. 1. Site location

Sampling and preparation

The measured sediment environmental parameters related to the availability of sedimentary metals are water temperature using a digital thermometer, salinity using a hand-refractometer, sediment Eh measurements were carried out *in situ* using a Hanna Instrument Eh meter (HI 8314). The probe used was a redox electrode (0–60°C), model IJ64, manufactured by Lonode, Australia. pH was measured using a Hanna Instrument pH meter (HI 8424) and concentration of dissolved oxygen using a DO meter.

Sample sediment was taken from the surface layer at approximately the oxic level with a depth of 1-3cm (estimated to be an area where there is still biological activity from marine biota) from the surface with an Ekman Grab Sampler. A 500g sediment sample was placed in polypropylene plastic. Polyethylene plastic had previously been soaked with 6 N HNO_3 and then rinsed with distilled water. The sediment samples were stored in a cool box at 4ºC. They were then placed on a tray, covered with tissues, and dried at room temperature. Subsequently, 5 grams of homogenized sediment were weighed and dried in an oven at 60ºC for 24 hours.

The sediment texture analysis was conducted following the guidelines established by the United Nations Environment Program **(UNEP, 1995)**. The total organic matter in the sediment was analyzed according to the standards set by the American Public Health Association, the American Water Works Association, and the Water Environment Federation (Part 5000 Aggregate Organic Constituents) **(APHA, 2005)**. Pb, Cr, and Hg metals were fractionated using the BCR Sequential Extraction method **(Sarkar *et al.*, 2014)**, which involves the analysis of four different binding phases.

Anadara granosa muscle was carefully taken and removed from the shell using a stainless-steel knife. Next, the sample was cleaned, rinsed, dried with paper and freeze-dried by lyophilization at 80ºC for 48 hours. The weight of *Anadara granosa* muscle was determined using a digital scale with a precision of 0.001g **(Samawi *et al*., 2020)**.

The concentration of metals in sediments and *Anadara granosa* was analyzed using an Atomic Absorption Spectrophotometer (Hitachi-Z 2000 Tandem Flame/Furnace AAS) with a limit of detection (LOD) for each metal as follows: Pb - 0.006ppm; Cr - 0.001ppm; Hg - 0.002ppm.

Data analysis

The data were analyzed descriptively in graphical form and to explain the relationship between environmental parameters with the concentration and fraction of metals in sediment and concentration in *Anadara granosa* at each research station using Principal Component Analysis (PCA).

RESULTS AND DISSCUSSION

1. Parameter of oceanography

The results of oceanographic parameter measurements in four stations include temperature, salinity, pH, dissolved oxygen (DO) (Table 1).

Table 1. Oceanographic parameters in four locations research

Station	Temperature (ºC)	Salinity (ppt)	pH	DO (mg/L)
Domestic harbor	29.4±0.35	29.3±0.61	7.77±0.12	5.9±0.68
Power plants area	28.3±0.31	30.44±1.39	6.83±0.49	6.3±0.21
Oil harbor	30.52±1.3	29.02±1.03	6.73±0.49	5.4±1.0
Gold mining	29.3±0.58	25.01±5.0	6.94±0.61	3.9±0.85

[1] Data primer 2024.

Table (1) shows that the water temperature of the research location ranges from 29.3 - 30.52°C, with a value that is classified as natural sea water. The salinity value, ranging from 25.01 to 30.44 ppt, suggests that the waters are influenced by freshwater from land, as natural seawater typically ranges from 34 to 35 ppt. The pH value, ranging from 6.73 to 7.77, indicates that the waters are slightly acidic, likely due to the influence of anthropogenic activities. The dissolved oxygen (DO) value ranges from 3.9 to 6.3 mg/L, with the lower values observed at the gold mining station, which is a semi-closed water system.

Sediment texture

The results of the sediment texture analysis at the research location are shown in Fig. (2).

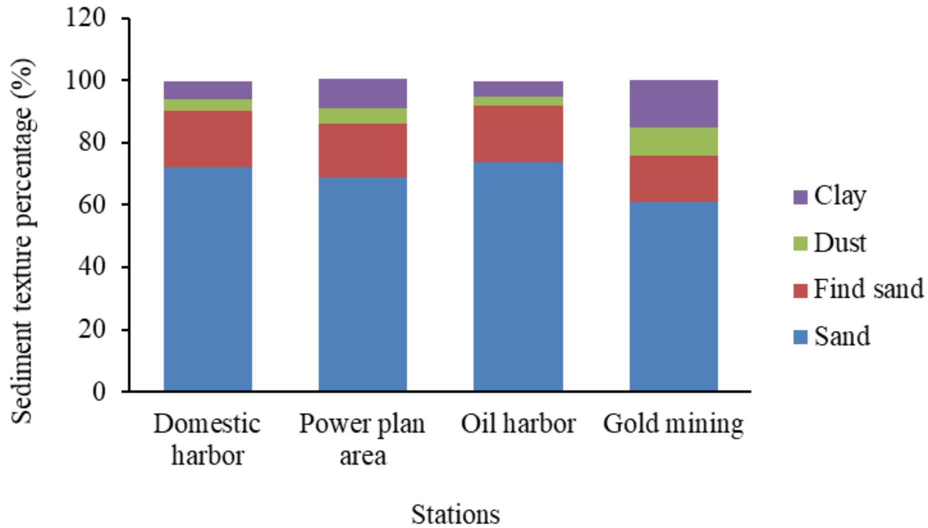

Fig. 2. Percentage fraction texture sediment (%)

Fig. (5) shows that the oil harbor has the highest sand composition due to its location in open waters and the influence of coastal currents. These currents trans harbor the smallest particles back to sea, while larger particles settle in coastal areas. This is in line with the findings of **Najamuddin *et al*., (2020)** which showed that the dominant sand texture in a location indicates strong water movement, which causes larger particles, such as sand, to settle on the seabed. The percentage of clay texture ranges from 5-15%. The high clay content at the gold mining site is due to the relatively calm bay conditions, where low current speeds result in the deposition of fine particles that are dominant in the sediment. Sand texture is closely related to the availability of metals in the sediment.

2. Total organic matter (TOM) in sediment

The total concentration of organic matter in sediment at four observation stations is presented in Fig. (3).

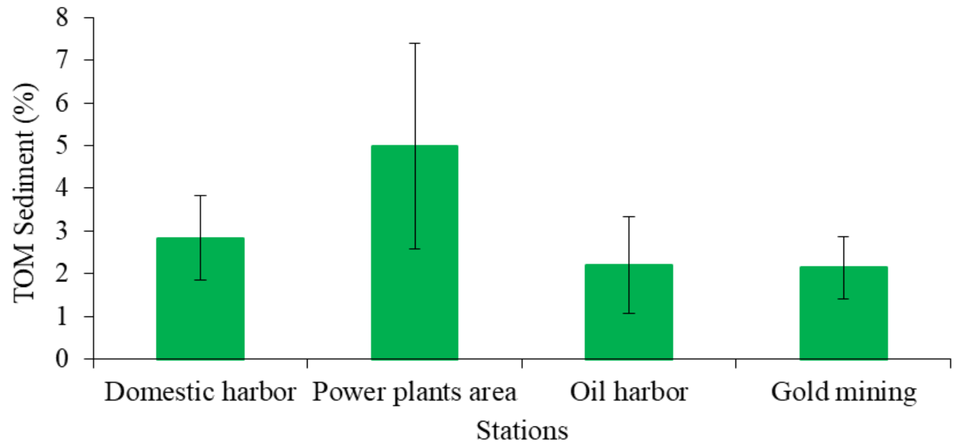

Fig. 3. Percentage of total organic matter in sediment

Fig. (3) shows the highest total organic matter concentration in the power plant area at 26% due to the presence of a dock where coal ships trans harbor coal. According to **Hartono *et al.* (2020)**, the power plant in Tidore uses low-quality coal, coal spills during transfer from ships to land cause the bottom of the harbor waters in the power plant area to be covered by coal. While in gold mining it is 6.97%. The high value at this location is because the water area is the estuary of the Taolas River and Tabobo River in Kao Bay. This is in accordance with the statement of **Dahuri *et al.* (1996)** that the river estuary is an area where water masses from land and sea meet which are still influenced by sea characteristics, such as salinity, tides, and seawater intrusion. In the upstream of this river estuary there are gold mining activities as well as community activities (settlements) and harbor that can contribute organic matter to the river estuary. High organic matter is closely related to the high availability of metals in marine sediments.

3. Redox potential (Eh) of sediment

The results of measuring the redox potential (Eh) values of marine sediments at four observation stations are presented in Fig. (4).

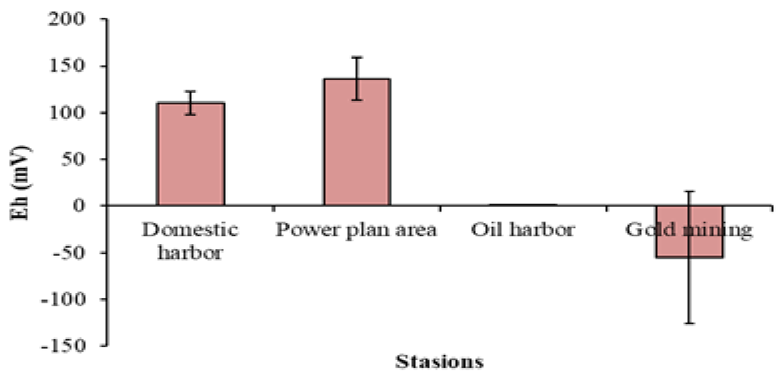

Fig. 4. Redox potential values (Eh) of sediments at four observation stations

The redox potential value of sediment in the observation location ranges from -55.083 to 136mV (Fig. 4), including the reduction and transition zone category or in aerobic/anaerobic conditions **(Odum, 1998)**. The location of domestic harbor, power plant areas and oil harbor are open harbor, so they are easily oxygenated. Open waters experience continuous water circulation so that the oxygen content increases, and ultimately increases the redox potential value **(Najamuddin, 2020)**. Positive redox potential values generally describe the characteristics of well-oxygenated sediments, including coarse sediments, or sediments that are poor in organic matter. Meanwhile, at the gold mining location, the negative redox potential value is a characteristic of sediments that are rich in organic matter and mostly consist of fine sediments. The condition of the sediment texture and organic matter content also greatly affect the Eh value. Sea waters with sandy sediments that are usually yellowish in color have positive Eh values, while soft sediments containing high organic matter have negative Eh values. Due to the very low organic matter content, the value of the sediment redox potential at the observation location is greatly influenced by the availability of dissolved oxygen. At the gold mining location and oil port, the Eh value is negative in line with the low dissolved oxygen content (Table 1 and Fig. 4). This is different from the location of the power plant port and domestic port with a higher Eh value due to oxidation events by dissolved oxygen. The Eh value is related to the bioavailability of metals in the sediment.

4. pH of Sediment

The results of sediment pH measurements at four observation stations are shown in Fig. (5).

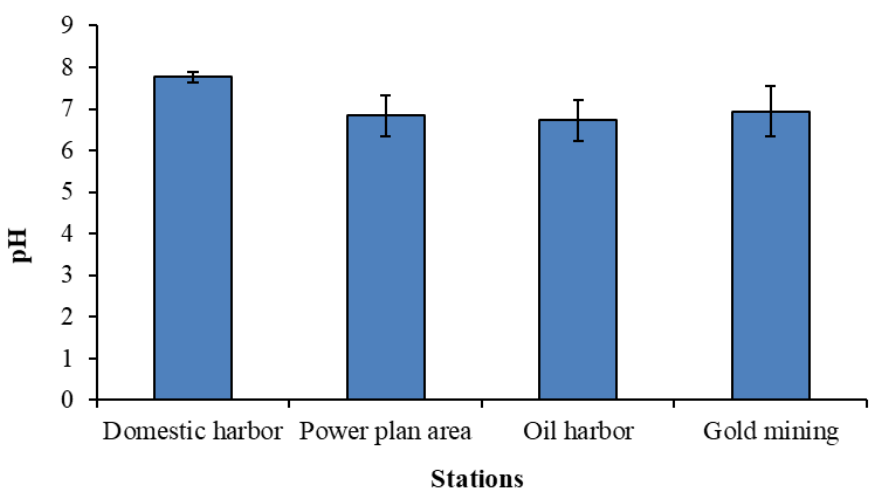

Fig. 5. Sediment pH values at four observation stations

Fig. (5) shows the low pH value of sediment at the gold mining site is directly prohibitional to the high percentage of dissolved organic matter there. This result is in line with **La Rowe (2020)**, who explained that the organic matter reduction process can affect sediment pH. Furthermore, **Hatje *et al*. (2003)** stated that pH can affect the adsorption value of metals (trace metals). Changes in pH affect adsorption because pH variations are linked to ion exchange mechanisms. This ion exchange process plays a key role in determining interactions both in solution and on the particle surface. The direct effect of pH on the adsorption rate is evident: at low pH (acidic), the adsorption rate is low, whereas at high pH (basic), the adsorption rate is higher. The increase in total organic matter (TOM) values in gold mining areas leads to a decrease in sediment pH and a lower redox potential (Eh-) due to reduced oxygen levels in the water. This reduction is caused by the decomposition of organic matter by microorganisms and the release of sulfuric acid **(LaRowe, 2020)**.

5. Total concentration of Pb, Cr, and Hg metal in sediment

Total concentration and fraction percentage Pb, Cr, and Hg metal results in sediment are presented in Table (2).

Table 2. Total concentration of Pb, Cr, and Hg metal in sediment at four stations

Stations	Total metal (mg/kg)		
	Pb	Cr	Hg
Domestic Harbor	15.11±1.42	0.092±0.034	0.0192±0.081
Power Plant	7.98±0.98	0.013±0.016	0.0028±0.003
Oil Harbor	14.37±1.34	0.317±0.253	0.0003±0.000
Gold Mining	9.02±2.38	0.141±0.053	0.0003±0.000

[2] data primer 2024.

Table (2) shows that the highest total Pb metal concentration values were found in domestic harbor, followed by oil harbor, gold mining areas, and the lowest in power plant areas. This trend can be attributed to the location of domestic harbor in Bastiong waters, which are the location of market activities, fish auctions, and harbor activities that produce organic waste. Both domestic harbor and oil harbor experience heavy trans harbor activities, which cause increased lead (Pb) pollution from anti-knock agents in fuel (e.g., tetraethyl Pb and tetramethyl Pb). This pollution is released into the atmosphere and then dissolve in water bodies **(Harmesa & Cordova, 2021)**. However, these values are still below the quality standards set by the **ANZEEC/ARMCANZ Guidelines (2000) and CCME (2001),** with the ANZECC/ARMCANZ guidelines indicating a low level of 50 and a high level of 220, and the CCME guidelines setting the ISQG at 30.2 and the PEL at 112. According to the US NOAA, the concentration of Pb in sediment is safe for biota at 46.7mg/ kg (ERL: Low Effect Range) and becomes hazardous at 218mg/ kg (ERM: Moderate Effect Range**) (Turki, 2007)**. The oil harbor is located in the Jambula waters, close to oil harbor and Kastela activities, tourist attractions, and tofu factories.

The main source of Cr metal here is organic materials. These values are also below the quality standards set by **NOAA (1996)**, **ANZEEC/ARMCANZ** guidelines

(2000) and **CCME (2001)**. The ANZECC/ARMCANZ guidelines for Cr are 80 (low) and 370 (high), and the CCME sets the ISQG at 52.3 and the PEL at 160. The US NOAA considers Cr concentrations in sediments to be safe for biota at 81mg/ kg (ERL) and hazardous at 370mg/ kg (ERM) **(Turkey, 2007)**. The highest Hg values were found in domestic harbor, followed by power plant areas, oil harbor, and gold mining areas. This suggests minimal anthropogenic sources, consistent with **Taylor (1964)**, who noted that natural Hg concentrations in the earth's crust average 0.08mg/ kg. The domestic area, near the active Gamalama volcano, is in line with **Darmono (2001)** statement that volcanic activity can release mercury. The power plant area, a coal-fired steam power plant, also showed Hg values related to its location. The low levels of Hg in the power plant were caused by the high aromatic content of coal and minimal metal particulates, as seen in the condensate waste.

6. Fractionation of Pb, Cr, and Hg metal in sediment

The results of fractionation of Pb, Cr and Hg metals in sediments in four locations in North Maluku waters are shown in Fig. (9).

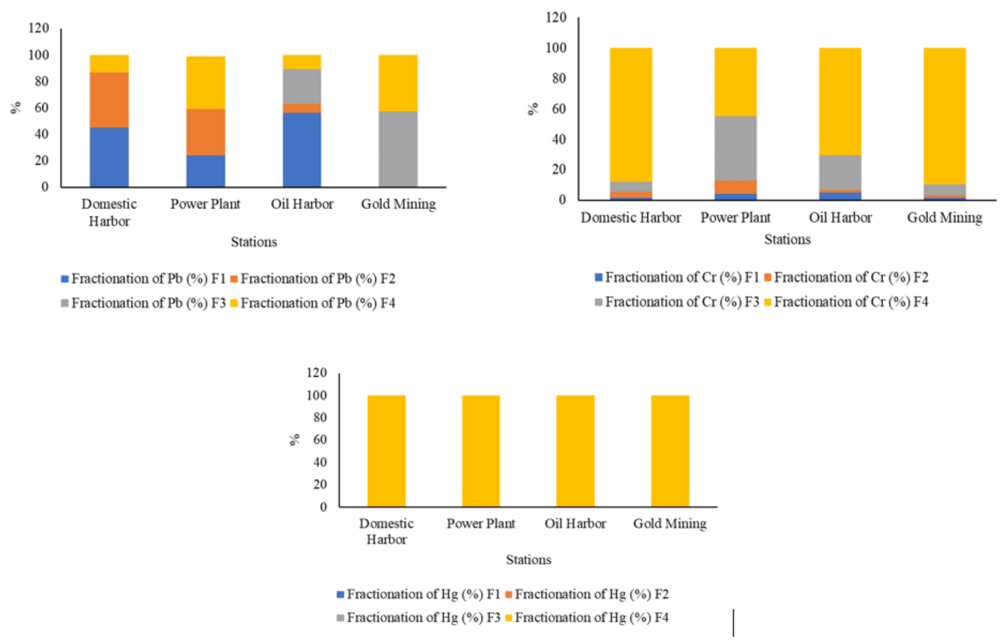

Fig. 6. Fraction percentage of Pb, Cr, and Hg metal in sediment

Fig. (6) shows the highest availability of Pb metal (F1,F2 dan F3) found in the power plants area, followed by domestic harbor, oil harbor, while in the gold mining area it is very low. The high F1 value in domestic harbor correlates with a low sediment pH value of 5.5, which is categorized as dangerous by **Lestari (2017)**. The higher F1 values in domestic harbor and oil harbor are in line with the high total metal values, similar to the findings in North Sumatra, where the Acid dissolved fraction (F1) in Fe is high along with the total metal content **(Yolanda *et al.*, 2019)**. The higher F1 value in oil harbor is due to its proximity to oil storage and tofu factory activities, both of which are sources of organic waste. The higher F1 value is also related to the low sediment pH value of 5.5 and Eh near the reduction zone of 0.863. High F2 values in domestic harbor, power plant areas, and oil harbor, combined with high redox potentials, reflect the metals found in fraction 2 associated with the concentration of Fe and Mn oxides in oxic sediments **(Luoma & Bryan 1981; Scouller *et al.*, 2006; Werorilangi 2012)**.

The concentration of Pb in Fractions 3 and 4 was found to be the highest in the gold mining location indicating low bioavailability of Pb with a TOM value of 2.199% with a low redox potential (reduction zone) and high F3 and F4 values indicating that Pb metal is mostly bound to sulfides, large organic molecules, and carbonates **(Guo *et al.*, 1997)**. This condition is in accordance with field observations where the color of the sediment is blackish brown in the mangrove-covered Kao Bay estuary.

Fractionation of Cr and Hg metals shows that they are dominated by fractions F3 and F4, this indicates low bioavailability of Cr and Hg metals because they are strongly bound even though F3 (fraction which can be oxidized) corresponds to the highest organic matter and redox potential values in the oxidative zone. These results are in line with the findings by **Ure *et al*. (1993)**, **Davidson *et al*. (1994)**, **Yuan *et al*. (2004)** and **Zhou *et al.* (2010)**, F3 (Oxidable Phase) indicates metals associated with organic matter and sulfides that can be released under oxidizing conditions (positive Eh value). The mobilization of metals in this fraction depends on the redox conditions, classifying it as a medium mobility fraction **(Najamuddin, 2017)**. High F4 represents the residual fraction (not bioavailable) which is widely found at gold mining sites. This fraction shows strong bonds between metals and sediment particles in the crystal structure of rock minerals, making it difficult to separate. According to **Tessier *et al*. (1979)**, **Werorilangi (2012)** and **Najamuddin (2017)**, oxidation conditions in natural waters can degrade organic matter, releasing dissolved metals, which causes high F3 values in sediments. High F1, F2, and F3 values in oil harbor highlight significant anthropogenic inputs polluting the waters. This observation is supported by **Werorilangi (2019)**, who stated that high labile fractions indicate pollution from anthropogenic sources, which are harmful to aquatic biota. Oil harbor in Jambula waters are heavily influenced by waste from oil reservoirs, tofu factories, and the Kastela tourist area, as well as fish farming as a source of organic matter.

7. Concentrations of Pb, Cr, and Hg metals in *Anadara granosa*

The Pb, Cr, and Hg content in *Anadara granosa* are shown in Fig. (7).

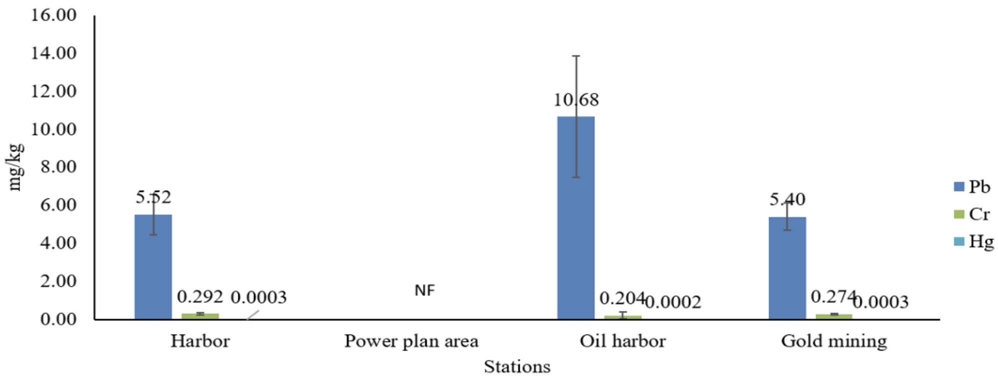

Fig. 7. Pb, Cr, and Hg content in *Anadara granosa* (NF = Not found)

Pb metal concentration in *Anadara granosa* in the research location 5.40-10.68mg/kg (Fig. 7). The greatest concentration of metals in shellfish was observed at the oil harbor, surpassing levels found in other locations. This value is higher than that found in *Anadara* sp. in South Sulawesi **(Samawi *et al.*, 2020)**. This elevated value of the labile fraction of Pb metal corresponds with the high concentration of the metal in *Anadara granosa*. The total value of the non-labile fraction indicates that the availability of Cr metal for *Anadara granosa* is minimal compared to the overall concentration of Cr metal in the sediment. Conversely, the low values of the total labile fraction at the domestic harbor, power plant area, and oil harbor stations suggest a lack of anthropogenic inputs that could contaminate the water. The results of Hg metal fractionation reveal that, at each station, the F4 (resistant) fraction exhibits a higher percentage than the F1, F2, and F3 (labile) fractions. This indicates that this fraction is non-bioavailable).

8. Relationship between environmental parameters and metal concentrations in sediment fractions and *Anadara granosa*

The results of principal component analysis (PCA) for Pb, Cr and Hg metal fractions can be seen in Figs. (8, 9 and 10).

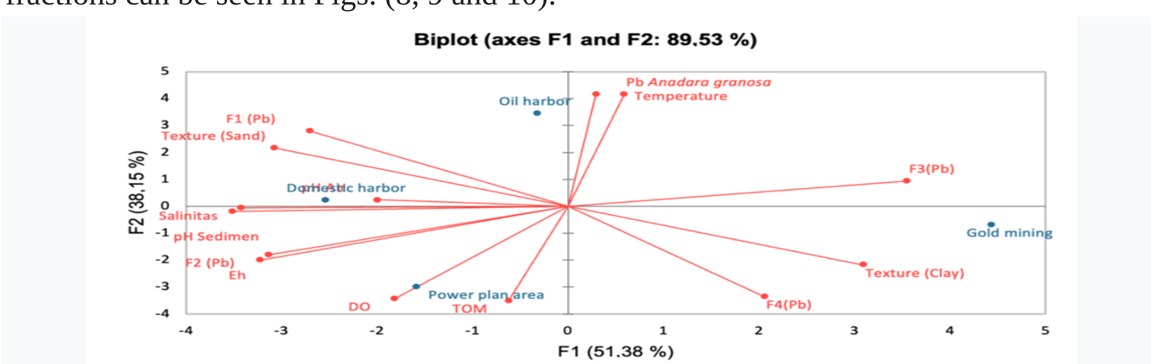

Fig. 8. Results of PCA analysis of environmental characteristics with metal fractionation Pb

Fraction 1 is characterized by a high percentage of sand texture and water pH, which was observed at Station 1. Fraction 2 is distinguished by redox potential entering the oxidative zone, low sediment pH, and high dissolved oxygen (DO) values, and is found in the power plant areas. Fraction 3 is characterized by high metal concentrations in biota and elevated water temperatures, which were observed in the oil harbor. Fraction 4 is characterized by a high percentage of clay texture, found in the gold mining area.

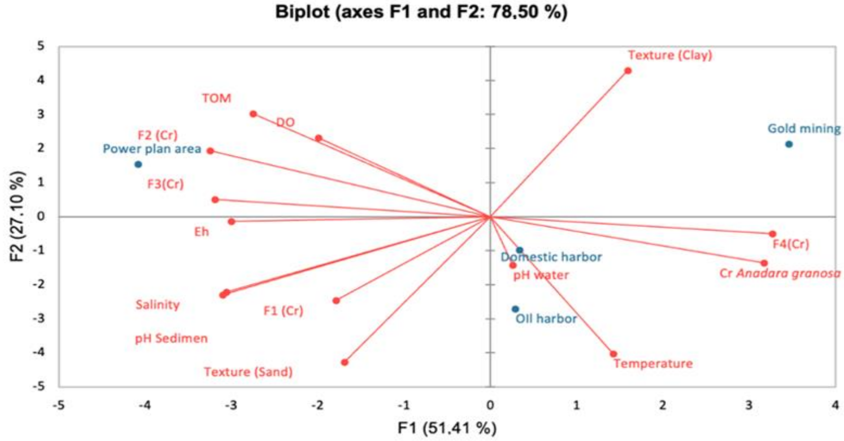

Fig. 9. Results of PCA analysis of environmental characteristics with metal fractionation Cr

Fig. (9) showed fraction 1 is characterized by high salinity values, redox potential values near the reduction zone, low sediment pH, and a high percentage of sand texture, as observed at the oil harbor. Fractions 2 and 3, found at the power plant area, are characterized by high dissolved oxygen (DO) levels, a large percentage of total organic matter (TOM), and redox potential values in the oxidative zone. Fraction 4, identified at the domestic harbor, is characterized by high water pH values and elevated metal concentrations in biota. Additionally, gold mining areas are characterized by a high clay texture.

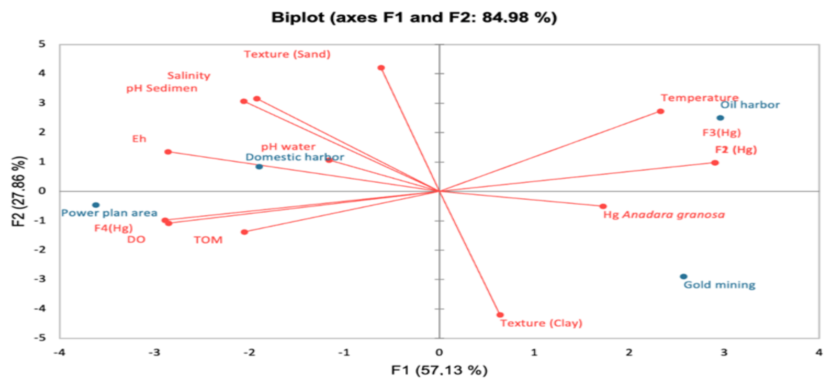

Fig. 10. Results of PCA analysis of environmental characteristics with metal fractionation Hg

Fig. (10) shows fractions 1, 2, and 3 are characterized by high water temperature values, observed at the oil harbor. Fraction 4, identified at the power plant area, is characterized by high percentages of total organic matter (TOM) and dissolved oxygen (DO). The domestic harbor exhibits high water pH values, redox potential (Eh) values in the oxidative zone, high salinity, low sediment pH, and a high percentage of sand texture. Gold mining areas are characterized by elevated metal concentrations in biota and a high percentage of clay texture.

Based on research results, it is shown that there has been an accumulation of Pb, Cr and Hg metals in sediments and *Anadara granosa*, it is necessary to provide waste water management installations and remediation processes at stations that have been contaminated with metals.

CONCLUSION

High total concentrations of Pb metal in sediments were found at all stations followed by Cr and Hg metals, but the concentrations were relatively low. Meanwhile, the high percentages of F1, F2 and F3 in Pb and Cr metals illustrate that these metals mostly come from anthropogenic sources and are bioavailable for *Anadara granosa*. Hg metal in sediments with a percentage of F4 exceeding the values of F1, F2, and F3 and the low concentration of Hg in *Anadara granosa* indicate that the bioavailability of Hg for biota is very low compared to the concentrations of other metals in sediments. Anthropogenic harbor activities provide greater bioavailability of metals Pb, Cr and Hg in sediments than gold mining in North Maluku Province.

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REFERENCES

APHA. (2005) American Public Health Association, American Water Works Association, Water Environment Federation.. Part 5000. Aggregate organic constituents. Organic carbon. In Standard methods for the examination of water and waste water. 21st Edition. Washington DC (US): American Public Health Association. 525p.

Australian and New Zealand Environment and Conservation Council (ANZECC) and Agriculture and Resource Management Council of Australia and New Zealand (ARMCANZ). (2000). Australian and New Zealand guidelines for fresh and marine water quality. Volume 1, Australian and New Zealand Environment and Conservation Council. Canberra. 29p.

Bastami, K.D.; M.R. Neyestani; N. Molamohyedin; E. Shafeian; S. Haghparast; I.A. Shirzadi and M. Bani Amam. (2018). Bioavailability, Mobility, and Origination of Metals in Sediments from Anzali Wetland, Caspian Sea. Marine Pollution Bulletin., 136: 22-32. doi: 10.1016/j.marpolbul.2018.08.059

Bubb, J. M. and J. N. Lester. (1991). The impact of heavy metals on lowland rivers and the implications for man and the environment. Sci. Total Environ., 100:207-233.

Canadian Council of Ministers of the Environment. (1999). Canadian sediment quality guidelines for the protection of aquatic life: merucy. In: Canadian Environmental Quality Guidelines. 5p.

Dahuri, R.; J. Rais; Ginting, S.P. and Sitepu, M.J. (1996). Integrated Management of Coastal and Ocean Resources. Pradnya Paramita., Jakarta, 305 pp.

Darmono. (2001). Environment and Pollution. Relation to Toxicology of Metal Compounds. University of Indonesia, Jakarta.

Davidson, C.M.; Thomas, P.P.; McVey, S.E; Perala, R.; Litlejohn, D. and Ure, A.M. (1994). Evaluation of a sequential extraction procedure for the speciation of heavy metals in sediments. Analy Chemi. Acta., 291: 277-286.

Guo, T.; DeLaune, R.D.; Patrick Jr, W.H. (1997). The influence of sediment redox chemistry on chemically active forms of arsenic, cadmium, chromium, and zinc in estuarine sediment. Environment International., 23 (3) : 305-316.

Harmesa and Cordova, M. R. (2021). A Preliminary Study on Heavy Metal Pollutants Chrome (Cr), Cadmium (Cd), and Lead (Pb) in Sediments and Beach Morning Glory Vegetation (Ipomoea pes-caprae) from Dasun Estuary, Rembang, Indonesia. Marine Pollution Bulletin., 162. 111819. https://doi.org/10.1016/j.marpolbul.2020.111819

Hartono, R. and A. Seng. (2020). Analysis of Low Rank Coal Fuel Efficiency in the Stoker Boiler of PLTU Tidore Unit 2. Journal of Mechanical Engineering., 5 (2).24-29.

Luoma, S.N. and Rainbow, P.S. (1981). A statistical assessment of the form of trace metals in oxidized estuarine sediments employing chemical extractants. Sci. Total Environ., 17: 165-196.

Lestari. (2017). Sequential Extraction for Heavy Metal Speciation in Sediments. Oceana Journal., 11(4): 1-12.

Najamuddin; Prartono T. and Nurjaya, W. (2016). Distribution and Behavior Of Dissolved and Particulate Pb and Zn In Jeneberang Estuary, Makassar, Jurnal Ilmu dan Teknologi Kelautan Tropis.. 8(1):11-28

Najamuddin.; Irmalita, T.; Rustam E. P. and Inayah. (2020). Influence of Sediment Characteristics on the Distribution and Accumulation of Heavy Metals Pb and Zn in River, Estuary and Beach Waters. Tropical Marine Journal., 23(1):1-14 doi: 10.14710/jkt.v23i1.5315.

National Oceanic and Atmospheric Administration (NOAA). (1996). Contaminants in Aquatic Habitats at Hazardous Waste Sites: Mercur. December Seattle, Washington.

Odum E.P. (1998). Basics of Ecology. Yogyakarta (ID): Gadjah Mada University Press.

LaRowe DE; S. Arndt; Bradley, J.A.; Estes, E.R.; Hoarfrost, A.; Lang, S.Q.; Lloyd, K.G.; Mahmoudi, N.; Orsi, W.D.; Shah Walter, S.R.; Steen, A.D.; Zhao, R.. (2020). The fate of organic carbon in marine sediments – New insights from recent data and analysis., 204. https://doi.org/10.1016/j.earscirev.2020.103146

Samawi M.F.; S. Werorilangi; R. Isyrini; Hendra. (2020). Bioavailability exchangeable phase of heavy metals in sediments and contamination in shellfish at estuaries on the west coast of South Sulawesi, Indonesia. AACL Bioflux., 13(4):2365-2374.

Sarkar, S. K.; Favas. P.J.C.; Rakhsit D. and Satpathy, K.K. (2014). Geochemical Speciation and Risk Assessment of Heavy Metals in Soils and Sediments. Environmental Risk Assessment of Soil Contamination. Chapter 25. doi: 10.5772/57295.

Scouller, R.C.; Snape, I.; J.S Stark and D.B. Gore. (2006). Evaluation of Geochemical methods for discrimination of metal contamination in Antarctic marine sediments. A case study from Cassy Stadion Chemosphere., 65:294-309

Situmorang S. P.; Sanusi, H.S. and Zainal Arifin. (2010). Geochemistry of heavy metals (Pb, Cr and Cu) in sediments and their potential availability in Benthic Biota in Berau Delta Waters, East Kalimantan. Journal of Marine Science., 2: 415-425.

Taylor, S.R. (1964). Abundance of chemical elements in the continental crust: a new table. Geochimica Cosmochimica Acta., 28(8):1273-1285.

Tessier A; P.G.C. Campbell and Bisson, M. (1979). Sequential extraction procedure for the speciation of particulate trace metals. Anal Chem., 51: 844-851.

Turki, A.J. (2007). Metal Speciation (Cd, Cu, Pb and Zn) in Sediments from Al Shabab Lagoon, Jeddah, Saudi Arabia. Marine Science., 18:191-210. doi: 10.4197/mar.18-1.112007

[UNEP] United Nations Environment Programme. (1995). Manual for the Geochemical Analyses of Marine Sediments and Suspended Particulate Matter. Reference Methods for Marine Pollution Studies No. 63. New York (US): Regional Seas.

Ure, A.M.; Quevauviller, F.; Muntau, H. and B. Griepink. (1993). Speciation of heavy metals in solids and harmonization of extraction techniques undertaken under the auspices of the BCR of the Commisiion of the European Communities. Int. J. of Environ. Anal. Chem., 51:135-151

Werorilangi, S. (2012). Speciation of bioavailable metals for benthic biota and spatial distribution patterns in coastal sediments in Makassar City. Dissertation. Agricultural Science Study Program. Hasanuddin University Postgraduate Program, Makassar.

Werorilangi, S.; Noor. A.; Samawi, M.F. and Ahmad, F. (2019). Spatial distribution of Pb, Cd, Cu, Zn metals and geochemical fractions in Makassar City Coastal Water sediments. Spermonde Journal of Marine Science., 5 (1):21-28.

Yolanda, D.S.; Prartono, T.; Koropitan, A.F.; Hartanto, M.T; Lestari, L.; Lubis, M.R.L (2019). Chemical Partition of Cu and Fe in Surface Sediments on the East and West Coasts of North Sumatera., 11(2):387-397.

Yuan, C.; J. Shi, B. He; J. Liu; L. Liang and G. Jiang. (2004). Speciation of heavy metals in marine sediments from the East China Sea by ICP-MS with sequential extraction. Environment International., 30:769-783.

Zhou, Y.; B. Zhao; Y. Peng and G. Chen. (2010). Influence of Mangrove Reforestation on Heavy Metal Accumulation and Speciation in Intertidal Sediments. Marine Pollution Bulletin., 60 : 1319–1324