

Potential for Heavy Metal Contamination in the Brown Macroalgae *Padina australis* from the Kayoa Islands, South Halmahera

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Received: May, 8, 2025/Accepted: December, 4, 2025

doi:10.24252/al-kimia.v13i2.56702

Abstract: Brown algae (*Padina australis*) are one of the macroalgae that have the ability to accumulate heavy metals, making them a bioindicator of environmental pollution levels. The waters of the Kayoa Islands, South Halmahera, are part of an area known as the world's coral triangle. This study aims to determine the levels of heavy metals Cd, Pb, and Hg accumulated in the brown algae *Padina australis*. The research method used was a wet destruction process and the samples were analyzed using an Atomic Absorption Spectrophotometer (AAS). The results obtained in this study from three sampling locations, namely the South, North, and West directions of the Kayoa Archipelago, show that Cd concentrations were higher compared to Pb and Hg concentrations. The Cd concentration was 0.69 mg/kg in the South, 0.62 mg/kg in the North, and 0.37 mg/kg in the West. The Pb concentration was 0.62 mg/kg in the South, 0.55 mg/kg in the North, and 0.34 mg/kg in the West. Meanwhile, the Hg concentration was 0.11 mg/kg in the South, 0.095 mg/kg in the North, and 0.075 mg/kg in the West. The levels of heavy metals Cd, Pb, and Hg detected in the brown algae *Padina australis* were still below the threshold quality standards of SNI 7387:2009 and FAO/WHO (2004). However, these levels should remain a concern for the local government because the accumulation of heavy metals can persist in aquatic biota over time. The results of this study can serve as a guideline for the management and mitigation of heavy metal pollution in the coastal and marine environments of South Halmahera.

Keywords: *Padina australis*, Heavy Metals, Kayoa Island.

INTRODUCTION

Macroalgae are large eukaryotic organisms inhabiting marine and freshwater environments and represent a significant source of bioactive compounds. Algae are organisms classified within the Kingdom Protista, resembling plants and characterized by a thallus body structure. Unlike most terrestrial plants, macroalgae obtain nutrients through osmosis and lack true roots, leaves, or a vascular system (Cordova & Muhtadi, 2017; Farizky et al., 2022). Algae possess chlorophyll pigments, which enable them to perform photosynthesis. Based on their morphology and pigmentation, algae are classified into three major groups: Chlorophyta (green algae), Phaeophyta (brown algae), and Rhodophyta (red algae). Brown algae serve as decomposers, producers, and a nutritional resource within their aquatic ecosystem. They provide as a source of sustenance and habitat within the marine ecology. This category of macroalgae possesses significant economic potential as raw materials for the industrial and healthcare sectors (Awalia, 2017). This type of macroalgae not only sustains the integrity of marine ecosystems and offers refuge for diverse marine organisms but also possesses considerable commercial potential (Teheni, 2019).

Brown algae, including the genus *Padina*, have been widely recognized as effective bioindicators of heavy metal pollution in marine ecosystems. For example, studies conducted

in Bais Bay, Philippines, demonstrated that *Sargassum*, *Padina*, and *Turbinaria* species accumulated cadmium from surrounding waters, indicating their potential role in biomonitoring and phytoremediation (Ho & Bantoto-Kinamot, 2021). Similarly, *Padina australis* from the Kayoa Islands, South Halmahera, exhibited measurable levels of Cd, Pb, and Hg, highlighting its importance as a sensitive indicator of environmental contamination (Hidayati et al., 2022). These findings support the relevance of brown algae not only for ecological and economic purposes but also for environmental monitoring.

The long-term increase in anthropogenic runoff is assumed to worsen the condition of coastal and marine ecosystems. Understanding the characteristics of terrestrial runoff in coastal areas is important to address information gaps, particularly in identifying ecosystem health. The research location is the Kayoa Islands, South Halmahera, which cover an area of 17,780 km² and are part of the region known as the world's coral triangle, as the area contains more than 1,000 km² of coral reef ecosystems (Khadijah et al., 2021). This study aims to determine the potential contamination of heavy metals Pb, Cd, and Hg in the brown macroalgae *Padina australis* as a result of organic material runoff.

RESEARCH METHODS

Sampling Location. The determination of the research station positions during the observation was conducted using a Global Positioning System (GPS) (Table 1). Spatially, the research location in the coastal waters of Kayoa Island was divided into three domains, namely the South, North, and West directions (Figure 1). The sampling location in the southern part of Kayoa Island was at the speedboat harbor area, the northern part was at the PELNI harbor area, while the western part was located in a residential area. Brown algae *Padina australis* samples were collected within 50 m from the shoreline, where the algae were attached to coral substrates during low tide.

Table 1. Geographic position of the research sampling Location

Sampling Location	North Latitude	East Longitude
South of Kayoa Island	0°06'08.53	127°24'45.00
North of Kayoa Island	0°07'29.00	127°27'3.00
West of Kayoa Island	0°0'37.00	127°24'26.00

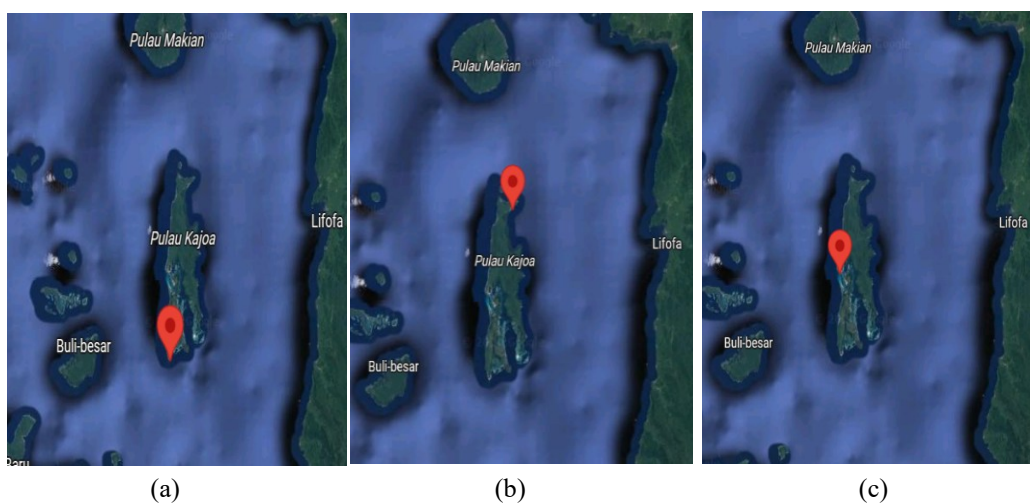


Figure 1. Sampling Location

(a). South ; (b). North ; (c). West

Materials and Tools

The tools used in this research included an analytical balance, a Spectra A50 Varian Atomic Absorption Spectrophotometer, a Sybron Thermolyne oven/incubator, a Sybron Thermolyne electric heater, a desiccator, beakers, a 100 mL volumetric flask, Erlenmeyer flasks, Whatman filter paper, a stirrer, and aluminum foil. The materials used were concentrated H_2O_2 , concentrated HNO_3 , and aquadest. In addition, water quality parameters were measured *in situ* using several instruments, including a refractometer to measure salinity and a thermometer to measure temperature (Rahmawati et al., 2020; Tampongangoy et al., 2021).

Measurements of temperature and salinity were conducted during each sampling event. Water clarity was measured using a Secchi disk by lowering it into the water until it was no longer visible. Current speed was determined by placing a floating object in the sea and measuring the distance and time of its displacement. A cork was used as the floating object.

Brown algae samples collected from the waters of Kayoa Island, weighing 1 kg, were washed with clean water and then sun-dried for one day until completely dry. The dried samples were subsequently cut into small pieces..

The analytical procedure employed a closed-system wet destruction method. Approximately 0.5 g of the prepared brown algae sample was weighed and subjected to wet destruction using a mixture of HNO_3 and H_2O_2 , with a volume ratio of 10 mL of HNO_3 and 2 mL of H_2O_2 . The digested samples were then analyzed using an Atomic Absorption Spectrophotometer (AAS)(Nuraini et al., 2019). The detailed procedures for heavy metal analysis followed the methods described in Table 2.

Table 2. Analysis of metal contamination data and its analytical methods

No.	Parameter	Unit	Method
1	Timbal (Pb)	mg/Kg	SNI 69892.46-2009
2	Cadmium (Cd)	mg/Kg	SNI 6989.16-2009
3	Mercury (Hg)	mg/Kg	SNI 2354.6-2016

RESULTS AND DISCUSSION

The research location was situated in the Kayoa Islands, with three sampling points established (Table 1). Brown algae samples were collected at a distance of 50 m from the shoreline. Of the three locations, the southern part of Kayoa Island was located at a speedboat base, the northern part was near the PELNI ship port, and the western part was located in a residential area. Two of the sampling locations function as ship ports. These two sampling locations are considered to contribute significantly to heavy metal contamination, as ship hull paint can be eroded by seawater and subsequently settle on the seabed. According to Risna et al. (2013), the use of ship hull paints and B3 chemicals (hazardous and toxic materials) in shipbuilding, which contain high metal concentrations, can cause deterioration of water quality. Furthermore to Bayram et al., (2013) stated that B3 chemicals can occur in the form of inorganic metal compounds or organic compounds.

Table 3 presents the average results of *in situ* water quality measurements conducted in the field. The parameters measured include temperature, salinity, turbidity, and current velocity.

Tabel 3. Results of in-situ water quality parameter measurements

No.	Water Quality Parameter	Coastal waters of Kayoa Island
1	Temperature	30,5 °C
2	Salinities	31,9 ‰
3	Brightness	45,2%
4	Velocity	7,6 cm/s

The water temperature in the waters of Kayoa Island was approximately 30.5 °C. Temperature influences the uptake of heavy metals by organisms as well as their metabolic rates. In addition, the toxicity of heavy metals such as Cd, Pb, and Hg, along with the distribution of organisms within an ecosystem, is affected by temperature (Hariyoto, 2017; Erniati et al., 2018; Putri et al., 2023). Temperature also affects the solubility processes of heavy metals entering marine environments. Higher temperatures can accelerate biodegradation processes carried out by aerobic decomposer bacteria, which may increase temperature and lead to the release of chemical substances into the environment (Putri et al., 2023). Surface water temperature conditions during the dry season are generally characterized by intense solar radiation, resulting in elevated water temperatures. The distribution of sea surface temperature in coastal waters is influenced by various meteorological factors, including rainfall, evaporation, humidity, air temperature, wind speed, and the intensity of solar radiation (Awalia, 2017; Setyaningrum et al., 2018).

The presence of heavy metals is also influenced by salinity in coastal and marine waters. The increase in the toxicity of heavy metals Cd, Pb, and Hg occurs due to the decrease in salinity, resulting in a higher level of heavy metal bioaccumulation (Hariyoto, 2017). The research results show that the salinity value of the Kayoa island is around 31.9 ‰. According to Waode et al. (2019), the salinity values of Indonesian waters generally range from 28-33‰. (Rustiah et al., 2020) states that differences in seawater salinity values can be caused by mixing due to ocean waves or the movement of water masses induced by wind.

TSS, brightness, and turbidity are parameters that are interrelated with each other. The increase in suspended solid concentration is proportional to the increase in turbidity concentration and inversely proportional to brightness. These three parameters play a very important role in the productivity of aquatic environments. This is closely related to the processes of photosynthesis and respiration of aquatic organisms. The presence of total suspended solids in the water affects the intensity of sunlight entering the water body (Rahmawati et al., 2020; Rustiah et al., 2019).

In this study, only brightness measurements were conducted. During the observation, the measurement results obtained a brightness value in the waters of the Kayoa island of around 45.2%. The brightness value of a body of water is closely related to the penetration of sunlight into the water body. Sunlight will aid in the process of photosynthesis, which will produce dissolved oxygen, an important factor in aquatic life (Cordova & Muhtadi, 2017). Furthermore, according to Tampongangoy et al. (2021), brightness affects algal growth; low brightness causes sunlight entering the water to influence the photosynthesis process.

Algae are impacted by velocity of sea water because they make it possible for the thallus to absorb, distribute, and supply the nutrients necessary for their growth (Farizky et al., 2022; Syahputra et al., 2023). The recorded current speed in the Kayoa island is 7.6 cm/s, which is deemed very mild, despite the presence of speedboat and PELNI ship ports to the north and south. Moreover, the comparatively feeble stream does not influence the accumulation of debris on the algae-covered coral reefs. The western region of the Kayoa island is a residential zone, however the current velocity is not excessively high. Typically, algae sample locations adjacent to populated areas encounter vigorous currents and elevated

wave activity, leading to the dispersion of community waste throughout the shoreline (Khadijah et al., 2021).

The community's heightened awareness deters them from fouling the beach. Bayram et al., (2013) asserts that the prevailing conditions will exhibit homogeneity, contingent upon the bathymetry and morphology of the shoreline. Wind stress on the sea surface generates wave heights, which subsequently produce surface currents. Increased wave height elevates current velocity, resulting in the formation of a robust longshore current that progressively induces beach erosion. Erosion occurs not just due to waves and currents but is also influenced by unstable bathymetric conditions.

Analysis of heavy metal content of Cd, Pb, and Hg in the brown algae Padina australis

Most macroalgae live in marine waters. To be able to grow, the macroalgae require a substrate to attach/live on. Epiphytic macroalgae on other objects such as stones, sandy stones, sandy soil, wood, mollusk shells, and epiphytes on other plants or different types of macroalgae (Hidayat et al., 2021). Brown algae are a type of macroalgae that grow wild in the waters of the Kayoa archipelago, and are one of the resources that grow on coral reefs. The morphological form of the brown algae *Padina australis* can be seen in Figure 2 below.



Figure 2. Brown Algae *Padina australis*

Identifying the sources of metals entering the waters is an important step in understanding the potential dangers of heavy metals. This allows us to determine whether the heavy metals originate from human activities or from natural sources. The bioavailability of metals in water is essential to support monitoring and evaluation activities of metal pollution in a region. Particulate of metals form will accumulate in the sediment at the bottom of the water or combine with particles suspended in the water column (Cordova & Muhtadi, 2017; Rustiah, et al., 2019). Because of this, the metals deposited in the sediment will be divided into various sediment fractions, which affects the ability of the metals to be utilized by aquatic organisms. In addition, it is important to determine the source of metals entering the waters, whether it comes from natural sources or from human activities (anthropogenic). The results of this identification are very helpful in monitoring and evaluating the level of metal pollution in certain areas (Sudir et al., 2017; Teheni, 2019).

From the analysis results of heavy metal content Cadmium (Cd), Lead (Pb), and Mercury (Hg) in the brown algae *Padina australis* samples using an Atomic Absorption Spectrophotometer at 3 sampling locations, presented in Table 4. The highest levels of the metal Cd compared to the metals Pb and Hg were found at 3 sampling locations in the waters of the Kayoa archipelago (Figure 3). The highest Cd concentration is in the South direction

at 0.69 mg/Kg, in the North direction at 0.62 mg/Kg, but the lowest in the West direction at 0.37 mg/Kg. The same goes for the Pb levels, where the highest concentration is in the Southern direction at 0.62 mg/Kg, while the sampling points in the Northern and Western directions are 0.55 mg/Kg and 0.34 mg/Kg, respectively.

Next, the concentration of Hg metal was also highest at the sampling point in the southern direction of the Kayoa archipelago, followed by the northern and western directions with concentrations of 0.11 mg/Kg, 0.095 mg/Kg, and 0.075 mg/Kg, respectively. The high levels of heavy metals Cd, Pb, and Hg in the southern direction are due to the location being a speed boat harbor, which is busy every day with the comings and goings of speed boats. Similarly, to the North is the location of the PELNI ship port, but the PELNI ships do not dock every day; there are specific schedules for the PELNI ships to dock at the Kayoa Islands port. The sources of Pb and Cd are likely from dock activities, fishermen's transportation, shipbuilding, wood preservatives, and anti-corrosive paint on the ship's hull. Ocean-going vessels operate in a highly corrosive environment, where seawater, salt, oxygen, and humidity are the main factors that accelerate the corrosion process. Meanwhile, at the sampling point in the West direction, the levels of heavy metals Cd, Pb, and Hg do not exceed the threshold, even though that point is a residential area (Khadijah et al., 2021; Rustiah et al., 2019).

Table 4. Results of heavy metal analysis in brown algae (*Padina australis*) along with a comparison of quality standards from SNI 7387:2009 and FAO/WHO, 2004

Location Point	Heavy Metals Type (mg/Kg)			Maximum Residue Limit (mg/Kg)		
	Cd	Pb	Hg	Cd	Pb	Hg
South direction	0,69	0,62	0,11	1,0	1,5	0,5
North direction	0,62	0,55	0,095	1,0	1,5	0,5
West direction	0,37	0,34	0,075	1,0	1,5	0,5

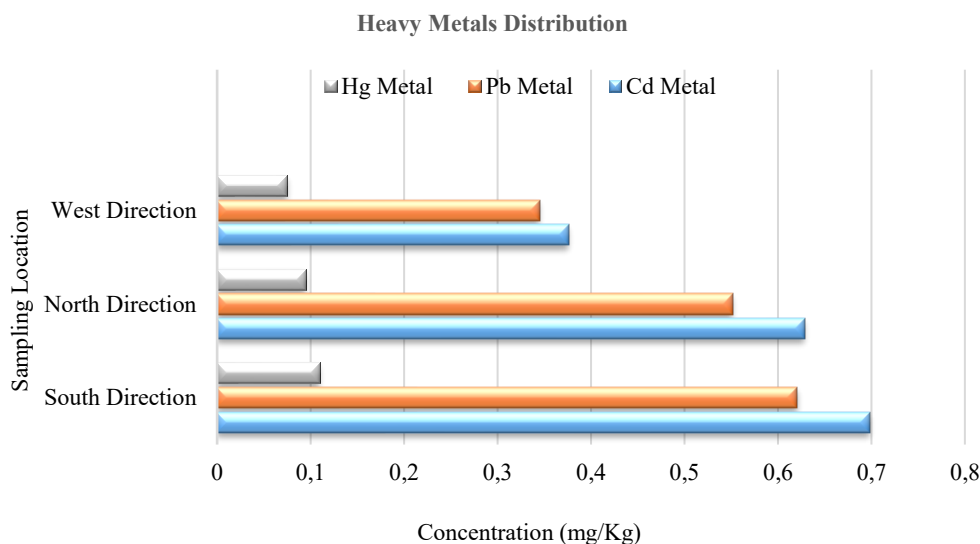


Figure 3. Comparison of heavy metals content distribution in the Kayoa Islands

The quality standard used in this study is SNI 7387:2009 for Cd and Pb metals, while for Hg metal it refers to the threshold set by FAO/WHO (2004). From the overall results of heavy metal analysis on brown algae (*Padina australis*) at all test points, they are still below the standard quality threshold. According to Siaka et al., (2016), the varying hydrodynamic conditions cause different levels of pollution to enter the waters of rivers, lakes, estuaries, and seas. Brown algae can produce functional compounds, especially agarophyte and carrageenophyte. The nutritional content of brown algae is usually very diverse due to various factors, including season, geographical location (habitat), species type, harvest age, and environmental conditions.

In addition, there are many causes of heavy metal accumulation in water bodies, one of which can originate from natural sources and human activities. Longer exposure can lead to the accumulation of heavy metals, especially cadmium (Cd), in the thallus of seaweed. If compared to pollution that originates from nature, pollution caused by humans has a greater impact. Maritime transportation that uses fuel and domestic waste, which can produce Cadmium (Cd) metal that can pollute seawater, is an example (Bhernama et al., 2021; Hidayat et al., 2021; Madsen et al., 2011; Rustiah et al., 2019).

The heavy metal content found in the waters has the potential to cause significant adverse effects on aquatic microorganisms. Bhernama et al., (2021) state that heavy metals have the property of easily binding to organic materials, then settling at the bottom of the water body. According to (Suryono & Djunaedi, 2017), Cd metal in aquatic environments is quickly absorbed by organisms in the form of free Cd^{2+} ions. Ions from seawater associate with Cd metal to form very strong complex compounds, such as CdCl_2 and CdCl^+ . Whereas Pb tends to accumulate in sediments through complexation processes with organic compounds. Beaches produce waste that can disrupt the life of organisms in the waters.

Due to the presence of Cadmium (Cd) in wastewater, the potential for environmental pollution is very high. The ability of biota to accumulate metals, or bioaccumulation, through the food chain allows the heavy metal Cd present in water bodies to accumulate. Therefore, hazardous substances are biologically metabolized and will impact organisms living in the water. So, high and low levels of pollution affect the amount of metals in the bodies of organisms (Fourqurean et al., 2012; Nur, 2013; Suryono & Djunaedi, 2017).

Mercury contamination in water bodies poses a significant danger to ecosystems and human survival. Mercury content can accumulate in the tissues of aquatic biota, thereby increasing the risk of mercury poisoning in humans who consume seafood from contaminated waters (Hidayat et al., 2021). Mercury pollution has been found in several waters in Indonesia. Based on the data obtained, one example of water contaminated with mercury waste is the Rupit River in Musi Rawas, located in South Sumatra, with a pollution index value of 2.17 mg/L, which is derived from the concentration of mercury found in the water and sediment of the river (Dewi, 2022). Additionally, the Cikaniki River located in West Java also experiences mercury pollution with a pollution index value of 2.05 mg/L, found in the sediment and water body of the river (Suryono & Djunaedi, 2017). Mercury pollution in aquatic biota was recorded in a journal by Nuraini (2017) in the Cikaniki River, where mercury content was found in fish, and a journal by (Cordova & Muhtadi, 2017; Yusuf et al., 2017) at several points in the river, where mercury content was found in green mussels. Mercury pollution in Indonesian waters has a significant impact on the environment. Mercury produced by various human activities can contaminate the surrounding water, air, and soil. Mercury in water can be converted into methylmercury ($\text{CH}_3\text{-Hg}$), which has a high level of

toxicity for aquatic organisms and humans (Hananingtyas, 2017). In addition, mercury pollution, especially in aquatic environments, can also affect aquatic ecosystems, causing disturbances to the natural balance and potentially reducing biodiversity (Awalia, 2017; Dunn et al., 2008). Mercury pollution has a very dangerous impact on human health (Hidayat et al., 2021; Vaeruzza et al., 2019).

CONCLUSIONS

Algae are autotrophic organisms and the largest contributors of oxygen in aquatic environments, capable of surviving in polluted water conditions. The accumulation of heavy metals Cd, Pb, and Hg occurs in the brown algae, *Padina australis*, which live in the waters of the Kayoa Islands, although their levels do not exceed the threshold (quality standard). Accumulation varies based on the type of metal and the time when the accumulation of Cd, Pb, and Hg occurs. The presence of algae serves as a biological parameter and can provide information to evaluate the condition, quality, and fertility of the water.

ACKNOWLEDGMENTS

An extraordinary thank you to all parties who have helped this research to be successfully carried out, and to the entire team who collaborated in writing this manuscript.

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