Heavy metal accumulation in the edible seaweeds *Caulerpa lentillifera* and *Caulerpa racemosa* from the coastal waters of Sanga-Sanga, Bongao, Tawi-Tawi, Philippines

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Abstract. Heavy metal contamination in marine ecosystems is a major environmental concern due to its persistence, toxicity, and bioaccumulative properties. This study assessed the accumulation of selected heavy metals in two edible seaweeds, *Caulerpa lentillifera* and *Caulerpa racemosa*, from the coastal waters of Sanga-Sanga, Bongao, Tawi-Tawi, Philippines. Samples were acid-digested and analyzed by atomic absorption spectrophotometry (AAS) with quality control procedures included blanks, replicates, and certified reference materials. Results revealed a consistent accumulation pattern of lead (Pb) > chromium (Cr) > cooper (Cu) > nickel (Ni) > cadmium (Cd) across replicates. *C. lentillifera* accumulated significantly higher concentrations of Pb (9.36 mg kg^{-1}), Cu (7.45 mg kg^{-1}), and Ni (2.37 mg kg^{-1}) than *C. racemosa* ($p < 0.05$) which is likely driven by its higher surface area-to-volume ratio and faster growth rate. This metal profile reflects an anthropogenic fingerprint dominated by Pb and Cr which is attributed to maritime activities and urban emissions near the Bongao port. Although concentrations comply with PNS/BAFS 194:2022 standards, Pb levels indicate a narrowing safety margin given their dietary importance. These findings establish baseline data for heavy metal bioaccumulation in *Caulerpa* species from the study area and support their continued safe consumption under current conditions. Furthermore, the results highlight the value of these seaweeds as bioindicators and underline the need for continued monitoring localized wastewater management to safeguard the long-term viability of the sea grape production in Tawi-Tawi.
Key Words: accumulation, food safety risk, environmental monitoring, trace element contamination, aquatic toxicology, dietary exposure.

Introduction. Contamination of heavy metal in marine ecosystems poses a significant global environmental challenge due to its persistence, toxicity, and bio accumulative properties, all of which pose substantial risks to biodiversity and ecosystem functioning (Masindi & Muedi 2018; Ali et al 2019; El-Sharkawy & Ali 2025). These contaminants originate from both natural processes and anthropogenic activities such as industrial discharge, mining, and urban runoff, which are evident in coastal environments. As heavy metals accumulate in sediments, they act as long-term environmental reservoirs that ultimately facilitate their entry into aquatic food webs. In rapidly developing regions such as Southeast Asia, the increasing coastal urbanization and industrialization have intensified heavy metal inputs, thereby heightening ecological risks in marine ecosystems (Ali et al 2019; Siddique et al 2025). These pollutants can bioaccumulate in marine organisms, including algae. Thus, posing risks to both ecosystem health and human consumers.

Edible macroalgae, particularly species of the genus *Caulerpa*, are widely recognized for their nutritional, ecological, and economic importance. *Caulerpa lentillifera* and *Caulerpa racemosa*, commonly referred to as 'sea grapes', are rich in essential nutrients and bioactive compounds including antioxidants and polyunsaturated fatty acids (Ismail et al 2020; Magdugo et al 2020; Syakilla et al 2022; Taslim et al 2024). Their high surface area and direct interaction with seawater enable them to efficiently absorb and accumulate dissolved substances, including heavy metals. Consequently, *Caulerpa* species are increasingly utilized as bioindicators of marine pollution due to their sensitivity to environmental changes and contaminant exposure (Zhou et al 2024).

In Southeast Asia, sea grapes are widely consumed as fresh vegetables and are valued both as a dietary component and as a source of livelihood in coastal communities. In the Philippines, these are locally known as "lato", "arosep", or "guto" and are commonly harvested and marketed for local consumption. Given their direct consumption and minimal processing involved, the accumulation of heavy metals in these edible seaweed raises important food safety concerns. Recent studies have shown that *Caulerpa* species can accumulate trace metals such as lead (Pb), and cadmium (Cd), particularly in areas influenced by human activities (Raza'I et al 2021; Deocaris et al 2022). This highlights the need to monitor contaminant levels to ensure consumer safety and sustainable utilization of these marine resources.

Despite the recognized importance of edible seaweeds and their potential to accumulate heavy metals, there is currently no available baseline data on the levels of heavy metal contamination in *Caulerpa* species from the coastal waters of Sanga-Sanga, Bongao, Tawi-Tawi, Philippines. This lack of localized information limits the assessment of potential risks associated with their consumption. Thus, this study aims to determine the concentrations of selected heavy metals in *C. lentillifera* and *C. racemosa*, including Pb, chromium (Cr), copper (Cu), nickel (Ni), and Cd. Furthermore, this study seeks to evaluate their implications for food safety and provide baseline data to support environmental monitoring and sustainable management of coastal resources in the region.

Material and Method. Fresh thalli of *C. lentillifera* and *C. racemosa* were collected from a single sampling station located in the Sanga-Sanga area, Bongao, Tawi-Tawi, Philippines (Figure 1). The site is characterized by shallow reef flats with sandy to muddy substrates where *Caulerpa* species form dense meadow population. Sampling was conducted through hand-harvesting during calm conditions by wading into shallow waters. A total of three biological replicates per species (n = 3) were collected from 5m apart within the same site to account for small-scale spatial variability. Extra care was taken to collect intact thalli while leaving the stolons and holdfasts undisturbed to allow natural regeneration. Collected samples were placed in clean polyethylene bags, transported to the laboratory under cooled conditions, and rinsed thoroughly with seawater followed by distilled water to remove adhering debris, epiphytes, and salts. The samples were oven-dried at 60-70°C until constant weight was achieved, then homogenized into a fine powder using a clean mortar and pestle. The powdered samples were stored in acid-washed containers to prevent contamination.

For digestion, approximately 0.5 g of powdered sample was treated with concentrated nitric acid (HNO_3, 65-70%) and hydrogen peroxide (H_2O_2, 30%) using a hotplate digestion method until a clear solution was obtained. Hydrochloric acid (HCl, 37%) was used for cleaning glassware and containers, while deionized water was employed for dilution and preparation of solutions. The digested samples were cooled, diluted to a fixed volume with deionized water, and transferred into acid-washed polyethylene vials for analysis.

The concentrations of Pb, Cr, Cu, Ni, and Cd were determined using atomic absorption spectrophotometry (AAS). Calibration curves were prepared using certified standard solutions prepared from analytical-grade reagents, and absorbance readings were taken at element-specific wavelengths. Quality assurance and control measures included the use of procedural blanks, replicate analyses, and certified reference materials (CRMs) to ensure accuracy and precision. All concentrations were expressed in mg kg^{-1} dry weight.

Descriptive statistics including mean and standard deviation (SD), were calculated for each metal concentration per species. Statistical analyses were conducted using Excel.

Measured concentrations were compared with available food safety guidelines for heavy metals. However, since Philippines regulatory limits are primarily established for seafood such as fish and shellfish, their applicability to macroalgae is limited. Therefore, international reference values for heavy metals in marine macroalgae and food safety guidelines (FAO/WHO Codex Alimentarius and European Commission (EC) for contaminants in food) and other recent literature were used as comparative benchmarks rather than absolute regulatory thresholds. Currently, there are no specific regulatory limits for heavy metals in edible seaweed. Thus, reinforcing the need for baseline assessment. This approach allows for a more appropriate assessment of potential health risks associated with the consumption of edible seaweed.

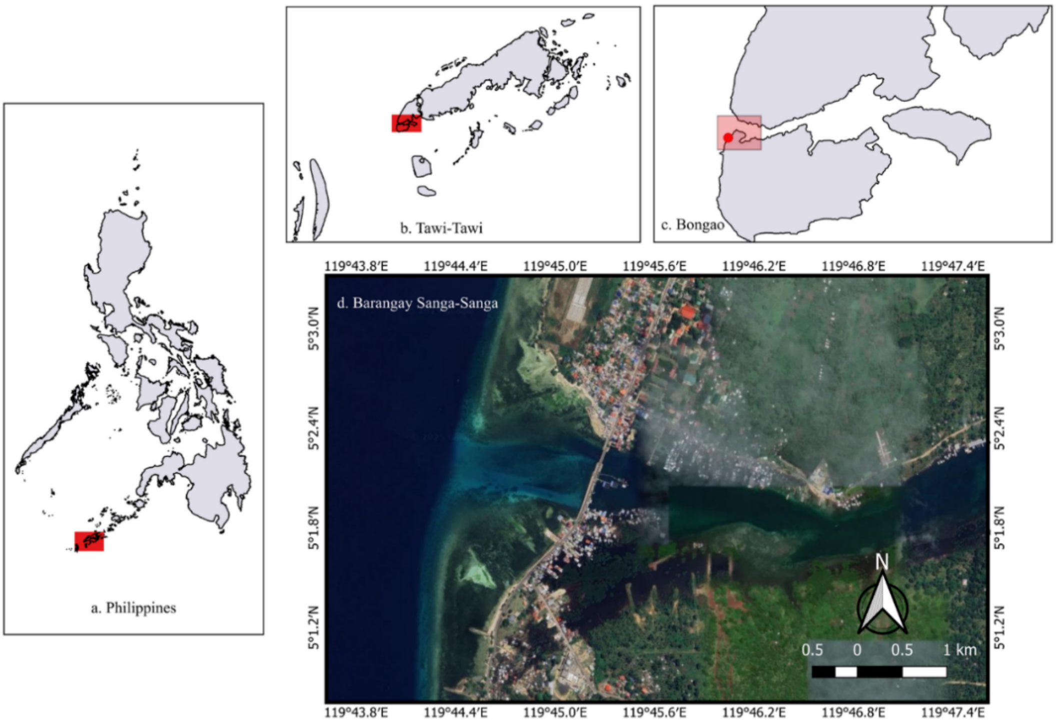

Figure 1. Map of the sampling site at Barangay Sanga-Sanga, Bongao, Tawi-Tawi.

Results and Discussion. The concentrations of Pb, Cr, Cu, Ni and Cd in *C. lentillifera* and *C. racemosa* are presented in Table 1. Across both species, Pb exhibited the highest concentration, followed by Cr, Cu, Ni and Cd, establishing a consistent accumulation pattern of Pb > Cr > Cu > Ni > Cd is observed across all examined replicates.

As shown in Table 1, statistical analysis revealed species-specific differences in metal accumulation. *C. lentillifera* accumulated significantly higher concentrations of Pb (9.36±0.17 mg kg^{-1}), Cu (7.4 ±0.09 mg kg^{-1}), and Ni (2.37±0.16 mg kg^{-1}) compared to *C. racemosa* (8.80±0.10, 6.51±0.19, and 1.69±0.11 mg kg^{-1}, respectively; $p < 0.05$). Cr showed no significant difference between species, while Cd levels, though slightly higher in *C. lentillifera*, were not statistically distinct ($p > 0.05$).

The differential accumulation profiles between the two species are likely driven by biological factors and environmental adaptability. The higher accumulation observed in *C. lentillifera* may be attributed to species-specific morphological and physiological traits, including its bead-like thalli and relatively higher surface area-to-volume ratio which enhance interaction with dissolved metals compared to the more slender and branched morphology of *C. racemosa*, thereby limiting its absorption capacity and resulting in comparatively lower concentrations (Delan et al 2015; Estrada et al 2020; Rakib et al 2021). Furthermore, frequently observed traits in *C. lentillifera* such as thinner ramuli and increased frond density are known to be correlated with higher sequestering efficiency for trace elements in varying habitats (Estrada et al 2020). Additionally, the relatively faster growth rate of *C. lentillifera* may further enhance metabolic activity and metal uptake efficiency (Uswatun et al 2025), reinforcing its role as a more sensitive bioindicator of subtle environmental changes, while both species remain effective monitors of coastal water quality.

Table 1

Heavy metal concentration in *Caulerpa* species from Sanga-Sanga, Bongao, Tawi-Tawi, Philippines

Heavy metal	*Caulerpa lentillifera (mg kg^{-1})*	*Caulerpa racemosa (mg kg^{-1})*	*Statistical significance ($p < 0.05$)*	*Interpretation*
Lead	9.36±0.17	8.80±0.10	Significant	Low to moderate level of accumulation
Chromium	8.61±0.19	8.31±0.07	Not significant	Low to moderate level of accumulation
Copper	7.45±0.09	6.51±0.19	Significant	Low level of accumulation
Nickel	2.37±0.16	1.69±0.11	Significant	Low level of accumulation
Cadmium	0.51±0.06	0.31±0.16	Not significant	Low to moderate level of accumulation

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Integrating these variables suggests that the measured metals did not act independently but rather exhibited a consistent accumulation pattern of Pb > Cr > Cu > Ni > Cd. This can be interpreted as a contamination fingerprint of the Sanga-Sanga/Bongao coastal zone where a stable uptake environment due to geochemical process and low-level anthropogenic inputs regulate metal availability (Pribadi et al 2025). This also reflects the combined influence of multiple sources and environmental processes rather than isolated inputs or natural geochemical weathering (Ali et al 2019). In typical aquatic systems, heavy metals often exist at their lowest concentrations in the water column but reach significant levels in coastal sediments, which then acts as a long-term reservoir for accumulation in marine organisms (Elvira et al 2020; Yunus et al 2020). The dominance of Pb and Cr suggests potential contributions from land-based runoff, domestic discharge and maritime activities which are commonly associated with coastal metal inputs (Ali et al 2019; Deocaris et al 2022; Murthy et al 2026). This dominance also indicates localized environmental pressure associated with human activities in the coastal zone such as near both docks and construction areas which have been known to exhibit elevated Cr and Pb levels due to intensive maritime traffic (Rashad et al 2024). Notably, the elevated Pb concentration (9.36 mg kg^{-1}) is substantially higher than values reported for *Caulerpa* in Manila Bay (2.71 mg kg^{-1}) and Vietnam's Van Phong Bay (0.58 mg kg^{-1}), which indicates a relatively stronger Pb signal in the study area (Perelonia et al 2017; Nguyen et al 2025). This elevated Pb signal, combined with elevated Cr and Cu levels can be pointed to a multi-source contamination profile stemming from the proximity of Bongao Port and the densely populated coastal communities. However, while total metal analysis provides a proxy for contamination, the human health risk is more accurately dictated by the bioaccessible fraction of these metals which is the portion released during digestion. In other tropical species, for example, Pb bioaccessbility has been recorded as relatively low (approximately 22-32%), which may mitigate some of the risks associated with the high total Pb concentrations observed here (Intawongse et al 2018). Consequently, the observed Pb > Cr > Cu pattern further supports the influence of maritime and urban sources, where the dominance of Pb may originate from land-based emissions from motor vehicle fumes and the widespread use of lead-acid batteries (Mahmoud et al 2024). Bongao is a high-traffic maritime and urban hub where the combustion of low-grade fuels and the improper disposal of transport-related batteries provide a constant flux of Pb into the costal fridge (Ray & Das 2023; Basri et al 2024). This finding is echoed in other Southeast Asian regions, where natural sea grapes collected from unmanage habitats show significantly higher Pb accumulation compared to those from controlled cultivation system, emphasizing the impact of localized environmental contamination (Purenji et al 2023). Meanwhile, Cu and Cr are associated with maritime activities including antifouling paints used on both wooden and steel vessels and corrosion of stainless-steel components and maritime coatings (Cunha et al 2024; Pribadi et al 2025; Murthy et al 2026;). Furthermore, the value of Cu and Cr concentrations recorded for *C. racemosa* (7.45 mg kg^{-1} and 8.61 mg kg^{-1}) and *C. lentillifera* (6.51 mg kg^{-1} and 8.31 mg kg^{-1}) exceeded the Philippine FDA threshold limits for health supplements of 2.0 mg kg^{-1} for Cu and 0.5 mg kg^{-1} for Cr (FDA 2025). Although this limit specifically targets processed supplements, it serves as a critical benchmark for assessing the safety of sea grapes as a functional food source in the local market, particularly in the absence of a specific Philippine National Standard for heavy metals in fresh macroalgae. Nevertheless, the Cu and Cr concentrations observed in this study remain lower than or comparable to levels reported in edible seaweeds from other countries (Makawita et al 2020; Premarathna et al 2022; Siddique et al 2025). This elevation may be linked to domestic waste inputs, including sewage discharge and plastic waste associated with coastal communities, though such interpretations should be considered indicative rather than definitive.

Comparing this fingerprint with other regions further supports its anthropogenic origin. For instance, *Caulerpa prolifera* in the restored Marchica lagoon of Morocco shows a hierarchy of Fe > Cr > Zn > Cu > Ni > Pb > Cd, where Pb is a minor component (Rahhou et al 2024). The reversal of this order in Sanga-Sanga, with Pb occupying the top position, highlights the influence of localized urbanization and maritime activity which may override natural geochemical signature (Khandaker et al 2021). Further regional contrasts are provided by research in Banten coast of Indonesia, where heavy metals were not detected in edible *Caulerpa* species which emphasizes that while the genus has high bioaccumulation potential, its metal burden is strictly regulated by environmental exposure (Sihono et al 2021). Furthermore, the fact that Pb levels are reaching 93.6% of the national permissible limit (10 mg kg^{-1}) suggests a narrowing safety margin, particularly in the coastal communities were *Caulerpa* is a dietary staple rather than an occasional supplement (Tahiluddin et al 2025). Given that heavy metals are non-biodegradable and persist in marine sediments, they can be remobilized during monsoon events or high-energy weather events, leading to potential increase of bioavailability and uptake in marine organisms (Pujari & Kapoor 2021; Edo et al 2024). This persistent nature means the coastal zone acts as a long-term reservoir for contaminants, which may be periodically released into the water column during periods of high environmental energy (Elvira et al 2020; Zhang et al 2023; Mali et al 2024).

A critical component of this study is the assessment of the harvested seaweeds against national and international food safety benchmarks. All measured concentrations currently fall within the permissible limits established by the PNS/BAFS 194:2022 General Standard for Contaminants and Toxins in Food and Feed, which adopts the maximum levels recommended by the Codex Alimentarius for aquatic products. Comparisons with international food safety benchmarks (e.g. FAO/WHO Codex Alimentarius) suggest that the detected concentrations pose minimal risk to human health under typical consumption levels (BAFS 2023). Comparable studies in the region have similarly reported low concentrations of heavy metals in *Caulerpa* species, supporting their continued use as edible marine resources under non-industrial conditions (Rakib et al 2021; Sarker et al 2021; Nguyen et al 2025). Nonetheless, continued monitoring is essential, particularly in light of increasing coastal development in the area and emerging environmental stressors such as microplastic-associated transport (Zhou et al 2024). However, the interpretation of these results requires caution. Rather than classifying the site as "non-polluted", the results are more appropriately interpreted as indicating relatively low levels of metal accumulation relative to highly industrialized hubs yet possessing a clear anthropogenic signature (Ajik & Tahiluddin 2023; Tume et al 2026). This cautious interpretation is necessary given that regulatory thresholds are not specifically established for macroalgae and that the study is based on a single sampling location. From a food safety perspective, the low heavy metal burden in both *Caulerpa* species supports their continued role as safe dietary resources, consistent with regional surveys of edible seaweeds (Perolonia et al 2016; Llanos & Dalawampu 2017). However, emerging concerns such as the synergistic effects of microplastics on Cd toxicity highlight the need for ongoing monitoring (Zhou et al 2024). Microplastics, which are pervasive in the coastal environment of Bongao, can act as vectors for Cd, increasing its potential for bioaccumulation and exacerbating its toxicological impact on the consumer (Zou et al 2025). Establishing this baseline is therefore critical not only for ecological risk assessment but also for guiding sustainable aquaculture practices and ensuring the long-term viability of the sea grape industry in Bongao.

The observed Pb-dominant fingerprint calls for the implementation of localized coastal management strategies to regulate waste discharge and protect this vital marine resource (Madjar & Scăețeanu 2025). While the seaweed remains safe for now, the encroachment of Pb and Cd levels toward regulatory thresholds suggests that unmonitored coastal development could quickly compromise the safety of these resources. In contrast, the relatively lower concentrations of Ni and Cd indicate limited influence from industrial or mining-related sources, consistent with the predominantly community-based setting of the study area and the absence of major industrial activities.

Similar accumulation patterns have been reported in Southeast Asian coastal systems, including Indonesia, Malaysia, and Vietnam, where Pb and Cr frequently dominate the metal spectrum in macroalgae due to mixed natural and anthropogenic inputs (Raza'i et al 2021; Nguyen et al 2025; Le & Long 2026). Furthermore, the overall pattern of relatively low metal concentrations aligns with the observations from less industrialized Southeast Asian coastal systems , where background levels are influenced more by natural processes than by intensive anthropogenic inputs (Cedeno et al 2019; Magdugo et al 2020; Yap & Al-Mutairi 2022; Jannat et al 2023). This consistency supports the interpretation that the observed pattern represents a characteristic signal of moderately impacted tropical coastal environments, where localized anthropogenic signals are present but remain within a moderate impact range compared to heavily urbanized coastal regions (Wong et al 2017). However, the absence of complementary sediment and water analyses limits definitive source attribution, and interpretations should therefore be considered indicative rather than conclusive.

Overall, the metal profile of *C. lentillifera* and *C. racemosa* from Sanga-Sanga, Bongao, Tawi-Tawi reveals a relatively low but detectable level of anthropogenic influence, rather than a non-impacted system. The balance between essential elements (Cu, Ni) and toxic metals (Pb, Cd), together with moderate concentration levels, indicates a functionally stable coastal ecosystem with emerging but manageable environmental pressure. Although the recorded concentrations of Cu and Cr exceeded the Philippine FDA limits established for health supplements, these values remain lower than or comparable to concentrations reported in edible seaweeds from other countries. These findings provide important baseline data for the region and support the continued use of *Caulerpa* species as bioindicators of coastal environmental quality. Furthermore, the results highlight the need for regular monitoring and localized management strategies to ensure the long-term safety and sustainability of edible seaweed resources in the area.

Conclusions. This study successfully establishes baseline data for heavy metal accumulation in *Caulerpa lentillifera* and *Caulerpa racemosa* harvested from Brgy. Sanga-Sanga, Bongao, Tawi-Tawi. The detected concentrations currently fall within permissible safety limits but indicate a narrowing safety margin, particularly for Pb. Overall metal levels remain low despite the presence of an anthropogenic signature, likely associated with intensified maritime activity and coastal urbanization. Although current concentrations pose minimal ecological and food safety risks, the potential for future contamination remains due to increasing residential density and coastal development near natural seaweed beds. The establishment of routine monitoring programs, localized wastewater management, and sustainable harvesting practices is strongly recommended to safeguard the long-term viability of sea grape cultivation as both a local food source and an emerging export commodity. These findings provide a critical baseline for future environmental assessments and support the sustainable management of coastal resources in Bongao, Tawi-Tawi.

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Conflict of Interest. The authors declare that there is no conflict of interest.

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