Research Article

The Concentrations of Selected Heavy Metals in the Surface Sediments in Three Locations of Southern Philippines

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ABSTRACT

Some heavy metals may be nutritionally essential, potentially beneficial, or toxic to life. Yet, frequent exposures to critical metals could also result in severe health and environmental impacts. This study aimed to investigate the coastal sediments of the three locations in the southern Philippines, namely: Oroquieta City, Baliangao, and Alicia, in terms of their concentrations in the following heavy metals: cobalt, chromium, copper, manganese, nickel, cadmium, and lead. The heavy metals in the surface sediment samples from the three locations were analyzed using AAS Perkin Elmer AA200. The results revealed that Oroquieta, Baliangao, and Alicia showed statistically equivalent low concentrations in cobalt, manganese, and lead, which were within the range of 21-29 ppm, 100-129 ppm, and 67-89 ppm, respectively. However, the three locations showed higher and statistically different concentrations of chromium, which was in the range of 672-831 ppm. The three locations also showed statistically different concentrations of copper, manganese, and lead. Of the three locations, nickel was only detected in Oroquieta, with a concentration of nearly 37 ppm. All the heavy metal concentrations, however, did not exceed the safety standards set by government agencies. In addition, there was no cadmium detected from the three locations. Hence, it can be concluded that the three coastal areas are very suitable for marine organisms like seashells and other marine organisms. The human impact has been fairly not alarming in the three sites.

Keywords: Heavy metals, Philippine coasts, Surface sediments, Surface sediments quality, Mindanao coasts.

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INTRODUCTION

The Philippines is situated in Southeast Asia, located in the Western Pacific Ocean. Its archipelago is composed of 7,641 islands. Being close to the equator and in the ring of fire, the Philippines is prone to earthquakes, landslides, and typhoons. However, it was considered one of the 17 mega-diverse countries[1] and is one of the countries with the highest biodiversity on the entire planet.[2] Philippine maritime waters are a habitat of inimitable and diverse marine life and an essential part of the Coral Triangle.[3]

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Oroquieta City, Baliangao Misamis Occidental, and Alicia Zamboanga Sibugay are three locations known for their vast supply of seashells. Baliangao and Alicia's seashell harvests per day are more than enough for its people and so, are sold across other regions. Hence, this study is considering these locations. This study was concerned with the concentrations of selected heavy metals (cobalt, chromium, copper, manganese, nickel, cadmium, and lead) in the surface sediments where clams and other seashells are gleaned. The heavy metal concentrations of the coastal sediments may serve as a gauging factor that may reflect the quality of the marine products harvested from the three locations. Heavy metals in this study were measured using the AAS Perkin Elmer AA200. Results of the three locations were compared among each other and significant differences were determined using Pearson and Tukey analysis. They were also compared to the standard guidelines set by authorized agencies that determined the present status of the coastal areas.

Heavy metal is defined based on its specific weight, but oftentimes the use of the term "heavy metal" for an element refers to pollution and toxicity.[4] Depending on the geological factors, metals and metalloids naturally occur in all ecosystems with varying concentrations. But anthropogenic activities alter their concentration which can be toxic at some level and may result in impairments in organisms' survival, reproduction, and behaviour.[5]

Copper finds its way to the seawater from human activities, like wood and fertilizer productions, and natural sources, like sea spray, forest fires, and decaying vegetation.[6] It is usually found in drinking water and would be beneficial at safe levels of less than two micrograms per litre.[7] Copper integrates with certain proteins to generate enzymes that act as catalysts to help several body functions and normal metabolic processes.[8] Cobalt in the soil is in the average concentration of 8 ppm.[7] The average cobalt concentration in marine sediments from 60 different sites globally was found to be 24.5 mg/kg, whereas the Pacific Ocean was found to have the highest concentration in the range of 38 mg/kg to 195 mg/kg.[9] As cobalt is carried by runoff, it mostly ends up in the ocean and sediments. Cobalt is essential when it accumulates in sediments, which is the habitat of the seashells because it improves the shells' quality.[10] High levels of nickel on sand and soils around the seashore damage aquatic life, especially the plants, including the reduction in the growth rate of algae. The accumulation of high levels of nickel in bivalves results in drastic impacts, including deformation of the shells.[11] Most manganese compounds have the tendency to bind to circulating particles or settle as sediments. Its concentration in soils is from 40 to 900 mg/kg, whereas natural waters, including lakes, rivers, and oceans, contain variable quantities of dissolved manganese, from 10 to 10,000 µg/L.[12] Manganese is essential for marine life and human use when used at safe levels of 2 micrograms per litre.[7] Lead is considered a metal with no beneficial effects on humans and animals.[4] Lead and its compounds are all considered toxic. This toxicity increases as its solubility increases. The more soluble the lead compound, the more toxic it becomes as it is taken by the body and accumulates over a prolonged period until a lethal quantity is achieved—a phenomenon known as cumulative poisoning.[13] Moreover, the lead collection in organs and shells of organisms in sand and sediments increases with exposure to high levels over time.[14] The shells are the leading storage site for heavy metals like lead. Chromium causes tumour formation, infertility, congenital disabilities, reduced capacity to fight diseases, and respiratory problems and is a threat to marine life because it destabilizes the ecosystem based on its toxicity.[15] Cadmium could travel long distances, especially as sludge. The sludge rich in cadmium ends up polluting the surface waters. Human consumption of cadmium and safe levels for marine life is less than 0.07 micrograms per litre, which is essential for human life when ingested as drinking water.[7]

MATERIALS AND METHODS

Sampling Scheme

The surface sediments being analyzed in this study were collected from the seashores of Oroquieta City; Baliangao, Misamis Occidental; Alicia, and Zamboanga Sibugay—all of which are located in the southern part of the Philippines. Figure 1 shows the locations of the sites on the Philippine map. Surface sediment samples were collected early in the morning from the intertidal zone up to shallow waters (0-10m). Amounts of sediments were collected from different spots in the coastal zone, which were then mixed to arrive at a composite sample for each location. Each composite sample was put in a polythene bag, labelled, placed in an icebox, and then

Figure 1: The Map of the Philippines Indicating the Three Sampling Sites in the Southern Part of Mindanao.

brought to the chemistry laboratories of the University of Science and Technology of Southern Philippines.

Data Collection Method

The sediment sample was poured into a clean sample pan and placed in an oven maintained at a temperature of 105°C for six hours. After such time, it was cooled and crushed using stainless steel mortar and pestle, sieved, and dried further at 105°C to be ready for analysis. About 0.5000 grams of finely pulverized sample was weighed for 30-min microwave digestion with 10 mL of aqua regia solution, then transferred to a 100-mL volumetric flask using a filtering funnel and then diluted with distilled water to the mark. After homogenization, metal concentration was determined using the atomic absorption spectrophotometer.

The results had to be evaluated against some standards. However, there are challenges met by the countries around the world in setting the quality guidelines--including the cost of the process and the insufficient data for correlating sediment analysis and its toxicity, and benthic invertebrate inhabitants' data both in coastal and river waters, which may be the basis for the derivation of standard guidelines. Because of this, studies in countries with no standard quality guidelines opt to compare their results to the guidelines set by other countries. This study compared its results to the guidelines set by the Hong Kong government, being the country closest to the Philippines with quality guidelines set for surface sediments.[16] The guidelines are presented in Table 1. The guidelines set two concentration standards - the ISQG-Low concentration (which is the trigger value) and the ISQG-High concentration. Below the ISQG-Low concentration, the probability of adverse effects is expected to be extremely low. While above the ISQG-High concentration, adverse biological effects are most likely to occur more frequently.[16]

One-way ANOVA determined any statistically significant differences among the measured values from Oroquieta City, Baliangao, and Alicia for the following heavy metals: copper, cobalt, manganese, lead, and chromium. Tukey comparison test was used to determine which pair of means were significantly different when one-way ANOVA indicated a significant difference.

Table 1: Hong Kong Sediments Quality Criteria.

Heavy metals	ISQG-Low concentration	ISQG-High concentration
Cadmium (Cd)	1.5	9.6
Chromium (Cr)	80	370
Copper (Cu)	65	270
Nickel (Ni)	40	
Lead (Pb)	75	218

Table 2: Concentrations of Heavy Metals in the Surface Sediments of Oroquieta, Baliangao, and Alicia.

Heavy Metal	Oroquieta City (ppm)	Baliangao (ppm)	Alicia (ppm)	Statistical Analysis
Copper	24.322	17.509	14.518	Significantly different
Cobalt	28.900	21.700	22.833	Not significantly different
Nickel	36.770	Not detected	Not detected	
Manganese	128.60	114.40	100.97	Significantly different
Lead	88.377	67.167	84.630	Significantly different
Chromium	829.40	672.20	831.00	Significantly different
Cadmium	Not detected	Not detected	Not detected	

RESULTS

The following Table 2 presents the concentrations of the heavy metals in the surface sediment samples from the three locations.

Tukey comparison tests revealed that at a 5% significant level, the three sites are significantly different in terms of their copper content. For manganese, Alicia and Oroquieta showed a significant difference, while Alicia-Baliangao and Oroquieta-Baliangao exhibited no significant differences. For both lead and chromium, Baliangao- Oroquieta, and Alicia-Baliangao were significantly different, while Alicia-Oroquieta were not.

Figure 2 shows the interval plots of the respective values for metal concentrations of the three samplings, which visibly revealed the comparison.

DISCUSSION

Surface sediments from Oroquieta City, Baliangao, and Alicia showed lower values for copper concentrations than the set values for ISQG-low by Hong Kong authorized agency. Oroquieta contains the highest concentration of said heavy metal. The three sampling sites do not have any metal production or mining. However, there are few lumber and milling businesses, especially in Oroquieta city, which had served the residents for building houses. Zamboanga Sibugay used to export logs, lumber, veneer, and plywood.

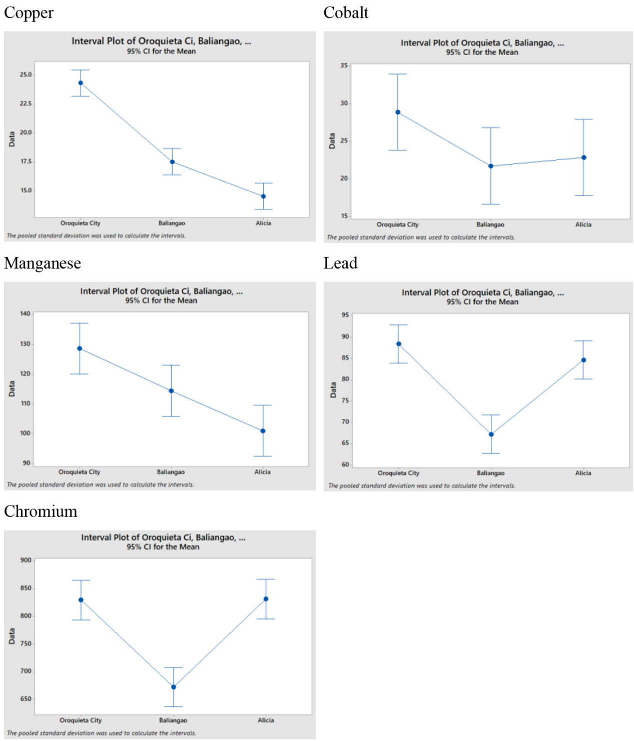

Figure 2: Interval Plot of the Heavy Metal Contents of the Surface Sediment Samples from Oroquieta City, Baliangao, and Alicia, Philippines.

These activities might have contributed to the higher accumulation of copper content of the two sites compared to Baliangao as these are identified sources of copper in seawater and surface sediments.[6] In addition, the sampling site in Oroquieta City is adjacent to a fish sanctuary where many mangroves are accumulating decaying organic matter and vegetation. This might have made Oroquieta city contain the highest copper content of the three sites.

Oroquieta is the only location with the detected nickel at a significantly low concentration. There is no nickel detected in the surface sediments from Baliangao and Alicia. Trash incinerators and power plants are the leading causes of nickel in the air and are bound in the acidic ground. Nickel appears more mobile and is always flushed to the groundwater. However, plants absorb nickel from contaminated seawater sources and are eventually consumed by humans through the food chain.[17] There were no waste incinerators or power plants in the said locations. Instead, mangroves and seagrasses can be observed in the sampling sites especially in Baliangao and Alicia with their respective huge fish sanctuaries nearby. For these reasons, nickel may not be detected in surface sediments.

For cobalt, the three locations showed lower concentrations, even lower than the mean range in the Pacific Ocean.[9] Human activities such as using and producing cobalt chemicals, processing ores that include cobalt, mining, vehicular exhaust, and coal combustion were the identified sources of cobalt. These sources eventually released cobalt into the atmosphere and found its way to the ocean.[18] This explained the significantly low concentration of cobalt in the surface sediments as there are not many of these human activities, except for vehicular exhaust.

Oroquieta has the highest concentration of manganese which showed no significant difference from Baliangao. Sources of manganese may be from mining, industries like the production of alloy and agrochemicals, welding, and other anthropogenic activities. There are not many of these activities in the three sites; hence, the researcher concluded that natural sources could be the most significant factor in such differences in the results. Natural sources of manganese include forest fires, ocean spray, vegetation, crustal rock, and volcanic activity.[12] Mount Malindang, a huge mountain range is located in Misamis Occidental, where Oroquieta and Baliangao are located.

For lead, the three locations have a slightly higher concentration compared to Hong Kong ISQG-low and far lower than the ISQG-high value for lead in surface sediments. Oroquieta City has the highest lead concentration in surface sediments which is not significantly different from Alicia's. Human activities, including combustion of solid waste, industrial processes, fuel combustion, and corrosion of leaded pipelines, are some of the causes of lead in seawater.[19] Of the three sites, Oroquieta and Alicia have ports for fishing vessels and ferryboats, respectively. This could be a reason for the high lead content in their respective surface sediments. In addition, Oroquieta City had been occupied for over a century; therefore, old pipelines might have deteriorated. Hence, Oroquieta had the highest lead concentration in surface sediments.

For chromium, surface sediments from Oroquieta City, Baliangao, and Alicia contain 124%, 82%, and 125% higher chromium concentrations than Hong Kong ISQG-high value, respectively. A higher chromium concentration could be from natural sources – from weathering rocks containing chromium and other natural occurrences. Sources of chromium in seawater are electroplating, leather tanning, textile industries, and cement production.[15] Of these possible sources from human activities, only cement production is present in the neighbouring city of Oroquieta. Hence, the researcher believes that the high chromium concentration of the sediments from the three locations is from natural sources. It could be from weathering rocks containing ores of mineral chromite that were leached or carried by runoff to the sea. Deposition of chromium might have been taking place long before. Domestic pollution can also be a possible cause of increased chromium concentration in the two locations. The study of He *et al*. (2020) on the identification of chromium in sediments in Xiaoqing River and Laizhou Bay found that high chromium was detected in the most polluted area of study.[20]

Earth's crust is comprised of about 0.1 ppm of cadmium.[21] The metal is in dry air or coated with the oxide in moist air. It is easily soluble in mineral acids. Since phosphate fertilizers are from mined rocks with varied cadmium contents, fertilizers contain a cadmium concentration of about 300 mg/kg. Hence, agricultural soils hold considerable cadmium concentrations.[22] However, cadmium in soil may be absorbed by crops such as rice.[23] For this reason, no cadmium was detected in the three locations as Mindanao is full of different vegetations and a lot of rice fields.

CONCLUSION

Therefore, these three locations--Oroquieta City, Baliangao, and Alicia-- are still clean, except for the high chromium content. The three coastal areas are suitable for marine organisms like seashells and other aquatic food products. The human impact has been relatively less in these three sites. However, there has been a hint of the anthropogenic impact caused by the increasing population and minor ports in Oroquieta and Alicia, as their lead and chromium contents appeared higher than in Baliangao. Although the high chromium concentrations of the three sites may be attributed to natural sources, which can be from weathering of rocks or from the soil itself, it still needs further study and analysis. In addition, the dense vegetation, the mangroves nearby, and seagrasses may have helped a lot in balancing the heavy metals in the surface sediments.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

ABBREVIATIONS

Cd: Cadmium; **Cr:** Chromium; **Cu:** Copper; **ISQG:** Interim Sediment Quality Guideline; **Ni:** Nickel; **Pb:** Lead; **Ppm:** parts per million.

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