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Analysis of Heavy Metals in Seawater Samples Collected from the Port of Ozamiz, Philippines

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Abstract

The rapid economic growth in Ozamiz City and escalating human activities in its port may pose a threat to the water quality in the area. As an agricultural area, runoff from pesticides and fertilizer residues may also put the marine waters at risk of heavy metal pollution. Urbanization may also contribute to the threat of water pollution. The marine waters in the Port of Ozamiz have not yet been investigated for possible heavy metal pollution. Hence, this study aimed to conduct an analysis of the heavy metals [magnesium (Mg), copper (Cu), cadmium (Cd), zinc (Zn), iron (Fe), manganese (Mn), mercury (Hg), lead (Pb)] in the seawater samples from the Port of Ozamiz. Temperature, pH, dissolved oxygen, nitrite, alkalinity, and hardness were the physicochemical characteristics of the seawater determined also in this study. Water samples were collected following the standard methods. Heavy metal analysis was carried out at the Department of Science and Technology laboratory in Cagayan de Oro City. Water analysis for the physicochemical parameters was done at the Bureau of Fisheries and Agriculture laboratory in Ozamiz City. Results showed that concentrations of Mg, Cu, Zn, and Cd are above the normal accepted values. All physicochemical parameters were within the desirable range except the water hardness. This study may provide baseline information for the local government of the city to formulate plans that could help reduce heavy metal pollution in the area.

Keywords: cadmium, copper, magnesium, pollution, zinc

Introduction

Heavy metal pollution of the sea is less than other types of water pollution, but its impacts on marine ecosystem and humans are very extensive. Aquatic organisms absorb the pollutants directly from water and indirectly from food chains (Khayatzadeh & Abbasi, 2010). Heavy metal contamination in seawater is becoming an increasingly severe threat to the naturally stressed marine ecosystem (Naser, 2013). In particular, heavy metal contamination in seawater near port areas has been reported (Lestari, 2004; Coban et al., 2009; Jonathan et al., 2011; Bozkurt et al., 2014; Tamilselvi et al., 2015; Mamon et al., 2016; Liu et al., 2017).

These heavy metals are naturally occurring. Natural weathering of rocks is a source of heavy metals (Nagajyoti et al., 2010). Volcanoes, sea sprays, and forest fires have also been reported to emit natural heavy metals (Vermette & Bingham, 1986; Seaward & Richardson, 1990; Ross, 1994). However, anthropogenic activities introduce them in excessive quantities in the environment which impose severe threats on the health of both humans and ecosystems (RoyChowdhury et al., 2018).

In the Philippines, few studies on heavy metal pollution in seawater were conducted. There were studies done in Manila Bay (Sia Su et al., 2009), Lingayen Gulf (Vinarao et al., 2014), East Bataan Coast (Cruz et al., 2015), and in coastal area in Misamis Oriental (Besagas at al., 2015) that showed presence of Cd, chromium (Cr), Cu, Pb, arsenic (Ar), and Hg. There were heavy metals that did not exceed the standard allowable limits, but their bioaccumulation in marine organisms might be likely to eventually pose a threat with prolonged exposure (Cruz et al., 2015).

In Northern Mindanao, Lacuna and Alviro (2014) were able to detect heavy metals in benthic foraminifera and nearshore sediments of Iligan City. Similar findings were found with the study of Ganaway and Lacuna (2014) in moderately polluted coasts of Iligan City. Iligan Bay connects with Panguil Bay on the southwestern part (Quiñones et al., 2002) which borders the province of Misamis Occidental. Panguil Bay is

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one of the priority fishery areas in Northern Mindanao. A recent study in the Port of Mukas, one of the ports that lies in Panguil Bay, shows high level of Pb (Jimenez et al., 2018). Ozamiz City is nestled at the entrance of the Panguil Bay with an excellent harbor location.

Ozamiz City is regarded as the center of commerce, transportation, health, and education. It is now emerging as one of the fastest growing cities in Northwestern Mindanao. Due to its transportation facilities, the volume of trade and commerce in recent years has tremendously increased. The primary means of transportation in the city is a motor tricycle, and its number has been increasing. Buses, vans, and jeepneys also provide long-distance transport for people and commodities. The Labo Airport in Ozamiz City as the only airport in Misamis Occidental serves flights from and to domestic destinations such as Cebu and Manila.

Recently, the construction of buildings (malls, food chains, hardware stores) roads, bridges, houses, banks, hospitals, and other infrastructures has been observed in Ozamiz City. More rural roads are also built to improve the transport service for agricultural products from remote lowlands and uplands. Gasoline stations even start to grow in number. The city is agricultural by resources. The rice fields and forest resources in Mt. Malindang have helped sustain the economy of the city. The Labo River that originates from Mt. Malindang provides water to lowland dwellers as well. However, this river is also the area for the quarry to provide construction materials for the city.

The rapid economic growth in Ozamiz City and escalating human activities in its port may pose a threat to the coastal environment. Also, as an agricultural area, runoff from pesticides and fertilizer residues may put the marine waters at risk of metal pollution. Quarrying and construction are also sources of heavy metals that can contaminate the coast. The port seawater has not yet been investigated for possible heavy metal contamination. Hence, an analysis of the heavy metals in seawater samples collected from the Port of Ozamiz was carried out. The physicochemical properties of the seawater were also determined. Results of this study may provide baseline information that could be useful for the local government of the city and other organizations in formulating plans that may help protect the marine ecosystem in the area.

Materials and Methods

Study area

This study was conducted at the Port of Ozamiz with coordinates 8.1449685° latitude and 123.84355° longitude. The port is nestled at the entrance of Panguil Bay (Figure 1). The Port of Ozamiz is one of the major seaports in the Philippines, that serves as the principal gateway port providing the shipping services for commodities and people across nearby places in Lanao del Norte and farther in Metro Manila, Cebu City, Dapitan, Iligan, Iloilo City, and Tagbilaran. About 160 meters from the port is the Cotta Beach where many residents in the city take their morning swim with the family especially during the weekend. Fishers also rely on seaweeds that they can harvest starting at about one kilometer away from the port.

Three sampling points were established in the study site. Sampling point 1 was at right side from the docking area of the harbor near human settlements. Recently, with the construction of Ozamiz Seaside Road, the waters are partitioned, and the houses are separated from the port area. The sampling point is also near some fish pens. Sampling point 2 is the docking area of the port. The port accommodates five shipping lines that provide service to passengers traveling to Metro Manila, Cebu City, Dapitan, Iligan, Iloilo City, Tagbilaran, and nearby places in Lanao del Norte. Seven ships and six ferry boats dock at the port. Sampling point 3 was at the left side of the docking area of the harbor near the Cotta Beach. This site is also near many houses that have been built along the coast near the beach.

Water sample collection

This study was carried out in August 2017. Permission from the Philippine Ports Authority (PPA) in Ozamiz City and the Department of Environment and Natural Resources (DENR) was obtained before the conduct of the research.

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Figure 1. **Satellite maps showing the Port of Ozamiz and locations of sampling points with actual pictures: (1) Sampling point 1 – right side of the docking area of the port near human settlements, (2) Sampling point 2 – docking area of the port, (3) Sampling point 3 – left side of the docking area of the port near the Cotta Beach. Source of maps: http://www.worldportsource.com/ports**

Water samples were collected using polyethylene bottles from a depth of 50.0 cm below the surface water following the method of Bozkurt et al. (2014). Three samples were taken as replicates for each sampling point. After rinsing the bottles three times with seawater, they were filled fully with seawater. The samples were stored in a box with ice for transfer to the Department of Science and Technology (DOST) laboratory in Cagayan de Oro City where water analysis for heavy metals was carried out and to the Bureau of Fisheries and Agriculture (BFAR) laboratory where the analysis of the physicochemcial parameters of the seawater was done. Magnesium, Cu, Cd, Zn, Fe, Mn, Hg, and Pb were the eight heavy metals analyzed. On-site determination of water temperature was carried out. The pH, dissolved oxygen (DO), nitrite, alkalinity, and water hardness were the physicochemical parameters analyzed at the BFAR laboratory.

Analysis of heavy metals and physicochemical parameters

At the DOST laboratory, the determination of mercury was carried out by DMA-80 thermal decomposition, amalgamation, and atomic absorption spectrophotometry (TDA ASS). Microwave-Assisted Digestion (Method 3030 A) and Direct Air-Acetylene Flame Method (Method 3111 B) were the standard methods used for all the other metals (Clesceri & American Public Health Association, 2005). Measurement was only done once.

The water quality guidelines and general effluent standards in the DENR Administrative Order No. 2016-08 (DENR, 2016) were used as the reference for the permissible limits of all heavy metals except for Mg. The ASEAN Marine Water Quality Management Guidelines and Monitoring Manual (ASEAN Secretariat, 2008) was used as the reference for the permissible limit of Mg because the value is not provided in the DENR guidelines. The ASEAN reference specifies that the standard limit for Mg can be applied to the marine waters of the Philippines. The Hach Water Test Kit (Test Kit Model FF3) was the test equipment used at the BFAR laboratory to measure the physicochemical

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parameters. The DENR and BFAR standards were used as the reference to determine the water quality in the Port of Ozamiz.

Results and Discussion

The mean concentrations of heavy metals in seawater samples collected from the Port of Ozamiz were found in the following order: Mg>Mn>Cu>Zn>Fe>Cd (Table 1). Concentrations of Mg, Cu, Zn, and Cd are above the permissible limits for coastal waters as prescribed by the DENR (2016) and ASEAN standards (ASEAN Secretariat, 2008). Iron and Mn were detected but their concentrations did not exceed the standards. Mercury and Pb were not detected in this study.

Table 1. Mean concentrations of heavy metals in seawater samples collected from the Port of Ozamiz.

Heavy Metals	Results (in ppm)	Standards (in ppm) (DENR, 2016 & ASEAN Secretariat, 2008)
Magnesium	*1262 ppm	1200
Copper	*0.10 ppm	0.02
Cadmium	*0.067 ppm	0.003
Zinc	*0.043 ppm	0.04
Iron	1.49 ppm	1.5
Manganese	0.10 ppm	0.4
Mercury	<MDL (MDL=0.000015 ppm)	-
Lead	<MDL (MDL=0.020 ppm)	-

Notes: MDL means Method Detection Limit
ppm - parts per million
*Above the normal accepted limit of the DENR

Some coastal areas in the Philippines are also contaminated with heavy metals due to anthropogenic activities by local and surrounding communities with high impact on marine biodiversity (Sia Su et al., 2009; Vinarao et al., 2014; Mamon et al., 2016). Mining, agricultural run-off, industrial and domestic effluents, brine discharges, mining, coastal modifications, oil pollution, sea traffic, port services, or bilge and ballast water disposals are sources or activities that introduce heavy metals to the coastal environment (Fu & Wang, 2011). The high influx of waste from toilets, kitchen, piggeries, laundry, and commercial establishments and indiscriminate dumping of solid waste into esteros that empty into the port seawater or Panguil Bay (Enguito et al., 2013) could also contribute to the high level of Mg, Zn, Cu, and Cd in the seawater samples collected from the Port of Ozamiz.

In this study, identification of point sources was not carried out, but the mere presence of the heavy metals that exceed the allowable limit requires serious attention from residents, government agencies, and non-government organizations. The concentrations of other metals may not have exceeded the limit, but these results could imply the possibility of this area to become more polluted without the strict implementation of appropriate measures and biomonitoring. Most marine ecosystems can cope with certain degree of heavy metal pollution, but severe pollution is reflected in the change of biotic communities that suffer the pollution (Förstner & Wittmann, 2012; Islam & Tanaka, 2004).

Magnesium ions in marine waters are essential for the normal growth and survival of phytoplankton (Solanki, 2012) and other marine organisms (Aruna & Felix, 2017) but elevated amount of this metal shows adverse effects on their survival due to bioaccumulation that results to toxicity (Vilhena et al., 2016; Diop et al., 2016; Srichandan et al., 2016; Gao et al., 2016; Monteiro et al., 2016). The high concentration of magnesium in this study could be attributed to fertilizer application taking into account that Ozamiz City is agricultural by resources. Magnesium in fertilizer serves as nutrients to plants for increased growth and yield (Cole et al., 2016) but excessive use of fertilizer results to heavy metal contamination in water (Sankhla et al., 2016). The indiscriminate dumping of plastic waste into esteros that empty into the coastal area (Enguito et al., 2013) may have also contributed to high level of Mg in the seawater since chemical industries nowadays add magnesium to plastics. During sampling, plastic wastes were seen floating in seawater near some coastal households.

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Copper is an essential element to organisms but is the most toxic heavy metal to marine biota above specific threshold level after Hg and silver (Ag) (Ansari et al., 2004). Bioaccumulation of Cu in marine organisms has been reported in several studies (Vilhena et al., 2016; Gao et al., 2016; Calbet et al., 2016; Bonanno & Di Martino, 2016; Vazquez-Luis et al., 2016). Exposure of marine organisms to Cu may elicit differential toxicological responses that affect metabolism (Zou et al., 2014).

In the Philippines, Velasquez et al. (2002) reported an elevated level of Cu near point sources in Manila Bay from anthropogenic input. In Iligan Bay, Cu had been detected, but species of foraminifera were still tolerant of this heavy metal (Lacuna & Alviro, 2014). However, the presence of Cu could imply strong potential of the area to become more polluted with continuous human-related activities that could lead to more Cu contamination. In this study, Cu could be introduced into the coastal waters through natural and anthropogenic activities. Runoff of pesticides and fungicides residues, influx of domestic waste, leaching of antifouling marine paints, and spill of leaded petrol from fishing boats could be the anthropogenic sources of Cu in the seawater. These sources of Cu contamination were also noted in the studies of Velasquez et al. (2002), Blossom (2007), and Brennecke et al. (2016) in marine waters.

Zinc is also an essential metal, but deleterious effects may occur with an excess of this heavy metal in marine ecosystem (Langston, 2017). Zinc is one of the most toxic metallic pollutants in marine waters (Nammalwar, 1983) when its concentration is elevated through human activities (Gale et al., 2004). Soluble Zn is far more readily absorbed and taken up by the tissues than the particulate Zn (Ansari et al., 2004). Bioaccumulation of Zn has been reported in planktons, algae, seagrass, bivalves, crustaceans, fishes, and mammals (Vilhena et al., 2016; Diop et al., 2016; Srichandan et al., 2016; Bonanno & Di Martino, 2016; Gao et al., 2016; Monteiro et al., 2016). Zinc contamination was also reported in Manila Bay suggesting anthropogenic inputs from intensive shipping activities in the area (Velasquez et al., 2002). In the Port of Ozamiz, the presence of Zn in the seawater above the threshold level could be due to electroplating activities in ships. A considerable amount of zinc leached from protection plate for boats containing the active zinc may contaminate the port seawater.

Cadmium is a nonessential metal and overloading of the assimilative capacity or threshold of the seawater could result in the occurrence of deleterious effects to marine biota (Langston, 2017). Pollution in the coastal waters of the world with Cd was reported in several studies (Velasquez et al., 2002; Bozkurt et al., 2014; Vinarao et al., 2014; Alharbi et al., 2017). This metal is toxic to organisms above a specific concentration and has adverse health effects in human metabolism (Sankhla et al., 2016). Bioaccumulation of Cd in marine organisms has been reported in the Philippines and other countries (Vinarao et al., 2014; Diop et al., 2016; Calbet et al., 2016; Srichandan et al., 2016; Vázquez-Luis et al., 2016; Gao et al., 2016; Gajdosechova et al., 2016).

Cadmium comes from natural sources, but industrial wastes, agricultural and mining activities have caused an increase in its level in marine waters (Ghani, 2015). Other possible sources of Cd are wastes disposed of batteries, Cd-containing alloys, foils, oils, automobile emissions and electrical equipment (Adokoh et al., 2011; An et al., 2010). Cadmium is also present as an impurity in phosphate fertilizers, detergents, and refined petroleum products (Mustapha & Lawal, 2014). In Ozamiz City, shipping activities, agricultural runoff, automobile emissions, and domestic wastes could be the sources of high level of Cd in the port area. Cadmium coatings provide vessels the good corrosion resistance (Wuana & Okieimen, 2011). The high influx of wastes from coastal households could be the sources of the high-level concentration of Cd in the port area of Ozamiz City.

The presence of Fe in this study may come from various anthropogenic sources such as agricultural products, domestic wastes, and shipping activities. Burridge et al. (2010) reported that feeds could be the source of Fe as they are formulated with various metals including Fe to fulfill complete mineral requirements for aquaculture. Some fish pens were observed in the study area that might contribute to the high concentration of Fe in the seawater in addition to Fe products

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released from nearby households and ships that may have contaminated the water.

As emphasized in the paper of Aguilar-Islas et al. (2016), iron availability influences algal community composition in coastal waters. Accordingly, sources of Fe in seawater could be sedimentary, fluvial, or aeolian. This metal is supposed to be found at exceedingly low concentrations due to its extremely low solubility in oxygenated seawater but it is quite high in this study although the quantity does not exceed the allowable limit.

Manganese is a new emerging contaminant in the environment (Pinsino et al., 2012). Municipal wastewater discharges, sewage sludge, alloys, steel, iron, ceramics, and fungicide products could be the contributing factors to the presence of manganese in this study. Many houses are built along the coast near the port, and septic tanks are absent. Sewage is drained directly into the seawater. This situation has to be looked at before the concentration of manganese as well as iron could rise above the allowable limit.

Table 2 shows the physicochemical properties of seawater in the Port of Ozamiz. The pH, DO, nitrite, and alkalinity are within the desirable range, but the mean value for hardness indicates that the seawater is very hard. The hardness of seawater is generally due to the presence of magnesium in high concentration.

Table 2. Physicochemical properties of seawater in the Port of Ozamiz.

Test Parameters	Sampling points			Mean	Desirable Range
	1	2	3		
Temperature (^{0}C)	27	28	29	28	25-31
pH	8.0	8.0	8.0	8.0	6.5-8.5
Dissolved oxygen (ppm)	8.5	11.0	11.3	10.27	5 (minimum)
Nitrite (ppm)	0.05	0.05	0.05	0.05	*<0.1
Alkalinity (mg/L)	159.0	115.0	116.00	130.00	*20-300
Hardness (mg/L)	4,270.0	4,290.0	4,410.0	4323.3	*Soft: 0-75 Moderately soft: 75-150 Hard: 150-300 Very hard: 300 up

*Based on BFAR standards

Water hardness, temperature, pH, and alkalinity are water quality characteristics affecting the bioavailability and toxicity of heavy metals to aquatic life (Davies et al., 1993). The degree of water hardness has the effect of reducing the toxicity of some metals such as cadmium and zinc through an antagonistic mechanism (Davies et al., 1993; Bradley & Sprague, 1985). Generally, metals become more toxic as temperature increases due to increased chemical activity and elevated metabolic rates of aquatic organisms (Davies, 1986). The water temperature in this study which is within the permissible range may have not facilitated toxicity with metals that are above the standard limit.

Increase in pH decreases the toxicity of heavy metals (Bradley & Sprague, 1985). Based on the result of this present study, the water pH is slightly basic which may also facilitate the solubilization of heavy metals. On the other hand, alkalinity primarily controls the bioavailability of heavy metals in natural waters (Davies et al., 1993). The alkalinity in this study is quite high but still within the allowable level. Increases in alkalinity also decrease heavy metal toxicity (Bradley & Sprague, 1985).

Nitrite is present in the studied samples but does not exceed the threshold level which may imply that the seawater is not highly polluted with effluents that are rich in ammonia which can lead to increased nitrite concentration in receiving waters. The slightly basic pH in the seawater of the Port of Ozamiz may also contribute to the solubilization of ammonia (Guštin & Marinšek-Logar, 2011).

The DO level in this study also indicates that the seawater is not highly polluted with organic matter, but monitoring of the quantity of waste drained into the seawater from nearby households or runoff is necessary. Oxygen consumption by microorganisms in the breakdown of organic matter present may result in oxygen sag if dumping of waste is not reduced or regulated. Discharge of untreated waste into near-shore waters could pose a threat to marine organisms as a result of pollution (Deepananda & Macusi, 2013).

The physicochemical characteristics of the seawater in the Port of Ozamiz may provide a baseline water profile that may affect heavy metal toxicity and bioavailability. Heavy metals become most harmful to aquatic life under conditions of low pH, low alkalinity, low DO, and elevated temperature. In this study, the water quality in the Port of Ozamiz may not have amplified the harmful effects of Mg, Cu, Zn, and Cd. However, the presence of these four heavy metals in the seawater of the Port of Ozamiz in high level calls for immediate attention before the coastal area becomes highly polluted that may pose health problems to the residents and threaten the aquatic life. Results of this study were already presented to the PPA, DENR and other stakeholders through conferences for research validation and dissemination.

Heavy metal pollution has become one of the most serious environmental issues. Contamination with heavy metals has increased dramatically in recent decades due to the natural occurrence or indiscriminate human activities that have altered the geochemical cycles and biochemical balance of these elements in the environment (Singh et al., 2011). In the marine environment, the persistence of heavy metals is regarded as one of the critical environmental concerns in developing countries (Factor & Chavez, 2012; Irnawati et al., 2014). The finding of this study is another proof that heavy metal pollution continues to occur in port area.

The toxicity and bioaccumulation characteristics of these heavy metals may cause the reduction in marine species and abundance (Ahmad et al., 2010; Hosono et al., 2011; Ramakritinan et al., 2012). Moreover, humans that rely on marine resources for food, industry, and recreation are at potential health risk (Naser, 2013). Heavy metals penetrate organisms via food, respiratory pathways or the skin (Jakimska et al., 2011). People observed swimming near the Port of Ozamiz could be at high risk of physical contact with the metals that pose a serious health threat. In humans, the biotoxic effects of heavy metals include developmental retardation, kidney damage, endocrine disruption, immunological dysfunction, neurological disorders, cancer, and other human health problems (Jaishankar et al., 2014; Mahurpawar, 2015).

Some of the heavy metals are essential trace elements that maintain metabolism but most of them are toxic to all life forms at high concentrations (Mohammed et al., 2011; Rao, 2014; Jan et al., 2015). However, the toxicity level of a few heavy metals to some organisms can be just above the permissible concentrations that are naturally present in the environment (Jaishankar et al., 2014). There are no known cases in Ozamiz City that manifest signs and symptoms related to heavy metals but actions are necessary to provide appropriate and effective solutions to heavy metal pollution in the Port of Ozamiz.

Conclusion and Recommendations

The concentrations of Mg, Cu, Zn, and Cd in the seawater samples collected from the Port of Ozamiz are above the minimum accepted limit. The physicochemical properties of the seawater are within the desirable range but the water is very hard.

Further study has to be conducted to determine the point sources of these heavy metals. Research on the bioaccumulation of heavy metals in fishes in the coastal area of Ozamiz City could provide additional information on the status of heavy metal pollution in the seawater. It is also recommended that the local government has to utilize the results of this study in formulating strategic plans to reduce the concentrations of these metals by full enforcement of the regulatory mechanisms that control the release of wastes from various point sources. There is also a need to review the existing policy related to recreation in the city to ensure that the residents are safe.

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