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RESEARCH PAPER

Analysis of heavy metals in seawater near a coal-fired power plant in a barangay of a city in the Philippine

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Abstract

The seawater in a barangay of a city in the Philippines close to a coal-fired power station is being used for recreational activities like swimming and source of seafood. This study was conducted to describe the physicochemical characteristics of the saltwater in April 2023, including its pH, temperature, and salinity. Additionally, the study detected and measured the levels of heavy metals in the collected saltwater, including cadmium, copper, iron, lead, and zinc. This study further interpreted the quantity of heavy metals present when compared to the allowable levels. Three (3) samples from two (2) points of the seawater of a barangay were collected during a normal tide with a depth of around two (2) meters. The sample sites were approximately ten (10) meters from the shore and thirty meters apart from each other. A Hanna Instrument pH meter, a mercuric-filled thermometer, and a refractometer were used to measure the physicochemical parameters such as pH, temperature, and salinity of the seawater respectively. Atomic Absorption Spectrophotometer (AAS) analysis was used to determine the heavy metals. The findings indicated that the seawater had a pH of 8.45±0.38, temperature of 30.00±1.10°C, and salinity of 30.55±0.27ppt. They were within the allowable level in April 2023. The quantities of copper (0.02±0.01 mg/L), iron (0.61±0.01 mg/L), and zinc (0.02±0.01 mg/L) were within allowable amounts. However, lead (1.44±0.19 mg/L) and cadmium (0.02±0.01 mg/L), were found beyond allowable of the permitted amounts. This study demonstrated the need for ongoing monitoring of the seawater around coal-fired power plants because of the beyond-allowable amounts of cadmium and lead.

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Introduction

Heavy metals are metallic chemical elements that are poisonous nervous, muscular system and the body (Singh *et al.*, 2022) and have a large atomic mass. By combining them with many environmental components such as water, soil, air, humans, and other living things that are exposed to the food chain, heavy metal toxicity can be further increased (Mitra *et al.*, 2022). Due to their environmental persistence and bioaccumulation, which prevents them from decomposing, these heavy metals enter the ecosystem through various chemicals and manufactured goods (Briffa *et al.*, 2020). Nowadays, marine contamination from coal-fired power plants is considered a significant environmental and public health issue, as the generation of electricity from coal combustion is still widely used (Hagemeyer *et al.*, 2019). Heavy metals liberated from coal ash that leak into the marine ecosystem due to improper disposal of coal combustion residuals cause harmful environmental effects that influence the health of humans (Kok *et al.*, 2019). Hence, the monitoring of heavy metals in areas near coal-fired power plants is relevant to evaluate their threat to the environment and human health (Canencia and Walag, 2016). Studying heavy metals aims to provide a clear explanation of their toxic consequences in living organisms, especially when highly exposed to humans through drinking or swimming, and their impact on the environment (Mitra *et al.*, 2022).

Among the most common heavy metals are cadmium, copper, iron, lead, mercury, and zinc. The majority of these heavy metals have accumulated as a result of human activity, primarily the combustion of coal and other fossil fuels. One of the heavy metals is lead. The skin can suffer negative effects from lead exposure in quantifiable doses. The most frequent symptoms are gingivitis, lead lines, and gingival brown pigmentation (Rerknimitr *et al.*, 2019). Lead's neurotoxic effects are especially pronounced in children, and even modest levels of exposure can cause considerable, and occasionally fatal, brain damage (Kragulj *et al.*, 2018). As a result, illnesses like mental retardation, renal system malignancy, and nervous system impairment manifest (Briffa *et al.*, 2020). Cadmium is another heavy metal that can damage the health of humans. Cadmium inhalation produces significant lung inflammation and damage, and prolonged exposure raises the risk of bone deterioration in adults, including osteoporosis. The accumulation of cadmium in a higher concentration is highly harmful to the kidney, resulting in kidney damage and renal failure (Engwa *et al.*, 2019). As cadmium does not undergo metabolic breakdown into less harmful and poorly excreted compounds, the human body has a limited ability to respond to it (Peana *et al.*, 2023). On the other hand, ingestion of high amounts of iron can interfere with cellular metabolism in the heart, liver, and central nervous system and cause direct corrosive damage to the gastrointestinal lining, resulting in nausea, vomiting, diarrhea, and abdominal pain. Hematemesis, perforation, and peritonitis can result from hemorrhagic necrosis of the gastrointestinal mucosa (Yuen, 2022). Zinc is another heavy metal. According to Briffa *et al.* (2020), zinc toxicity can have a variety of negative effects on the respiratory and gastrointestinal tracts, as well as the brain. It can cause nausea and vomiting by acting as an intestinal irritant. While copper, as a trace element, is needed at levels ranging from one to one hundred mg per day. Exposure to too much copper from environmental sources can result in copper toxicity (Royer *et al.*, 2022). Excess heavy metals in the human body like copper, zinc, and iron cause the development of free radicals, which can lead to serious issues such as mental impairment. Exceeding the maximum limit for exposure can be overall poisonous and seriously harmful to human health.

In a city in the Philippines, a coal-fired power plant is located on the seashore of a barangay. Recreational activities such as swimming and fishing are done by the residents on the coastline in that barangay. The coal-fired power plant that is near this barangay burns fossil fuels, and the smoke is released into the air, which carries these heavy metals and goes to the sea. Heavy metal beyond the allowable standard can pose a risk to human health. This study analyzed the presence of heavy metals in seawater near a coal-fired power plant in a city in the Philippines. Specifically, the study determined the physicochemical parameters including the pH, temperature, and salinity of the collected seawater in April 2023. The study also detected and quantified the presence of heavy metals such as cadmium, copper, iron, lead, and zinc in the collected seawater. Interpretation of the amount of cadmium, copper, iron, lead, and zinc present in the collected seawater when compared to the accepted values set by the Department of Environment and Natural Resources (DENR) of the Philippines was also done. This study established that during April 2023, the seawater in a barangay of a city in the Philippines near the coal-fired power plant had within the acceptable pH, temperature, and salinity. The cadmium and lead levels were beyond the acceptable level based on the Department of Environment and Natural Resources of the Philippines. The results are beneficial to the Department of Health (DOH), the Department of Environment and Natural Resources (DENR), the community, and future researchers as a basis for managing the beyond-acceptable levels of cadmium and lead.

Materials and methods

Physicochemical analysis: per Hydrogen (pH), temperature, and salinity of seawater

Treatment of containers for seawater sample collection

According to the study of Canencia and Walag (2016), the treatment of the sample containers was done two days before the collection of the seawater samples. Six polyethylene terephthalate (PET) bottles were washed with detergent and rinsed with water. Afterward, the containers were cleansed with distilled water. A 20 L mega box was likewise washed with detergent and rinsed with water. On the mega box, a ratio of 45 mL of ten percent nitric acid to five L of distilled water was prepared. The PET bottles were soaked in the mega box with the solution for 48 hours. After two days, the PET bottles were taken out of the box. An ample amount of the prepared solution was poured into a PET bottle, covered with its lid, and shaken to rinse the bottle. The solution was transferred to another PET bottle, covered, and shaken for rinsing. The steps were repeated for every bottle and allowed to air dry in a plastic tray.

Beakers and 50 mL and 100 mL volumetric flasks with covers were washed using dishwashing liquid and rinsed with water. The glassware was placed inside the megabox container with a prepared solution of 45 mL of ten percent nitric acid to five L of distilled water. The glasswares were allowed to soak for 24 hours. After 24 hours, the beakers and volumetric flasks with covers were swilled with the prepared solution. They were taken out of the container, cleansed with ultrapure water, and allowed to air dry in a tray.

During the seawater collection, six samples were taken using 1000 mL treated bottle containers. Three samples from two points were collected during a normal tide with a depth of around two meters. The sample sites were approximately ten meters from the shore and thirty meters apart from each other. The bottles were rinsed three times with seawater before they were filled to condition the bottles with the samples (Berx *et al.*, 2017). After which, each of the three bottles was filled with one liter of seawater from point A, and the other three bottles were filled with one L of seawater from point box for transfer to a research laboratory of a private university.

Determination of the physicochemical parameters

The physicochemical parameters determined in this study were the pH, temperature, and salinity of the seawater. For the test of pH, a Hanna Instrument pH meter was used. The beakers were soaked with ten percent nitric oxide and distilled water for 24 hours, and the electrode tip was rinsed with distilled water. Three replicates were tested for each point. A 20 mL sample was then placed on each beaker for point A and point B. Calibration was done using the 4.0 pH buffers. The electrode was allowed to stabilize, and the pH for both sampling sites A and B were observed and recorded. The electrode was rinsed with distilled water after each sample point to avoid sample carryover (Hanna Instrument Manual, 2018).

For a temperature test, a mercury-filled thermometer was used. The thermometer was immersed two-thirds in the seawater's surface, and the measurement was taken in the central flowing location. It remained immersed for one minute to adjust to the water temperature before removing it from the seawater. Testing was immediately done before the other tests to avoid interference from possible changes. The waiting duration for the result was one minute and was recorded in degrees Celsius.

For the salinity test, a refractometer was used. This tool depends on the refractive index to determine the level of water salinity. The refractometer was calibrated before adding one to three drops of the water sample using a pipette onto the prism. The plate was then closed, and the reading of the scale was seen through the eyepiece. The results were recorded in parts per thousand (Bakri *et al.*, 2020).

Heavy metal analysis

Sample preparation for heavy metal analysis

For the digestion of the seawater, the collected samples in the sample containers were allowed to be adjusted at room temperature. The bottles were inverted and shaken to resuspend the contents. A 100 mL of the seawater samples were pipetted out from point A and point B, respectively, and delivered to a 100 mL beaker. Five mL of nitric acid was then pipetted and delivered to each of the beakers with a 100 mL seawater sample, which was slowly boiled on a hot plate inside the fume hood until both beakers reached the 20 mL mark. This process lasted for about two hours. Both beakers were then removed from the hot plate and allowed to cool down on a clean surface. The digested point A sample was filtered through a Whatman filter paper that was inserted inside the mouth of the funnel, where a 100 mL volumetric flask was attached at the end to catch the filtrate. The remnants from the beaker were washed with acidified ultrapure water and filtered into the same filter container as Point A. The procedure for washing the remnants was done three times consecutively. The same procedure has been followed for the Point B sample.

Qualitative heavy metal analysis

For the analysis of cadmium, copper, iron, lead, and zinc, different standards were used, but the same procedures were followed for all five heavy metals.

Acidified ultrapure water was prepared using a millipore water purification system to ensure that there were no interferences in the AAS. A 16 mL of nitric acid was transferred to a 500 mL Duran Glass bottle using a serological pipette, and 500 mL of ultrapure water was added. The contents of the reagent bottle were mixed by inverting multiple times to obtain acidified ultrapure water. Lastly, the acidified ultrapure water was transferred into a wash bottle for easy transfer for each dilution.

For the preparation of the heavy metal standards, dilution was done. A 500 ppm of the standard solution was prepared from 1000 ppm using a serological pipette by delivering 50 mL of each heavy metal chemical in a labeled 100 mL volumetric flask and adding a small portion of the acidified ultrapure water to mix the contents properly and filling until the upper meniscus of the volumetric flask. From this, 20 mL was taken and transferred into another labeled 100 mL volumetric flask for the preparation of a 100 ppm solution, which was added with acidified ultrapure water until the upper meniscus. From that, 50 mL was taken and transferred into another labeled 100 mL volumetric flask to prepare a 50 ppm solution, which was also filled with the acidified pure water until the upper meniscus. The succeeding four volumetric flasks have a capacity of 50 mL in preparation for the five ppm, three ppm, one ppm, and 0.5 ppm solutions from the 50 ppm dilution to be utilized as the heavy metal standards in the AAS reading. After the exact amounts were added to each volumetric flask, a small portion of the acidified ultrapure water was added to mix the contents properly, and the volumetric flasks until the upper meniscus.

The prepared standards and the samples from point A and point B were readied to be introduced to the AAS for the identification and concentration determination of cadmium, copper, iron, lead, and zinc.

For the AAS to run, the fuel used was acetylene gas. The fuel gas regulator was adjusted to 12.5 kg/cm on the left side and 12 kg/cm on the right side.

Using the AAS for heavy metal analysis, the hollow cathode lamp was initially changed depending on the heavy metal to be analyzed. A computer that has programmed software called SpectrAA (Worksheet AA Software) was used to encode the needed information on heavy metals. As soon as the SpectrAA software was opened, the application was manipulated for the reading of the standards and sampling points in the order: of copper, lead, cadmium, zinc, and iron. The order was done at random, and there was no indication of this. A different worksheet was then created for each heavy metal and named based on the specific element that was run. The use of flame was manually chosen as the method type, and the monochromator was set at 284.3. All the tabular keys and sections that were not changed were already set by the software.

The labeled standards and concentrations were checked to ensure the encoded data was in the worksheet. Optimization of the hollow cathode lamp was done to reach more than the maximum amount the AAS can run for stable analysis.

A spare 100 mL volumetric flask of ultrapure water served as the blank sample for the AAS reading. This ensured no interference with the absorbance. While aspirating, the flame inside the AAS was blue, indicating that there were no contaminated substances in the blank sample. From this, the standards were enabled to calibrate the readings, and the samples were then introduced to the AAS. The reading of the samples from sampling points A and B turned the flame from blue to red, indicating the presence of substances that the AAS was aspirating. After readings of standards and samples, SpectrAA quantified the number of heavy metals in the seawater samples and presented the results.

Quantification heavy metals analysis

A standard calibration curve for each heavy metal within the analyte sample's range was created. The software SpectrAA was utilized to enable an automated quantification of heavy metals in seawater. The calibration curve was set to 0.5 ppm, one ppm, three ppm, and five ppm for all heavy metals. A linear calibration curve was achieved to define the range of concentrations of the heavy metals to be quantified at which the instrument responds linearly. Concentrations beyond the standard calibration curve, on the other hand, resulted in uncal or an unsuccessful run. All heavy metals follow the same procedure, but each utilizes different lamps specific for their detection and quantification. The concentration of the heavy metals and absorbance were then presented by the AAS system based on the standard calibration curve.

Interpretation of the amount of heavy metals in seawater

The interpretation of the heavy metals present in the seawater near the coal-fired power plant was based on the standard of the Department of Environment and Natural Resources of the Philippines.

Waste disposal

Proper waste disposal was strictly followed in this study. Excess acidified ultrapure water, together with the sample water that was obtained from the sample sites, was discarded on the sink while the water in the faucet was running. The heavy metal wastes were discarded in a waste container labeled “Organic Waste with Heavy Metals”. Used gloves were thrown into a waste container labeled “Used Gloves”. Pipette tips were discarded first in a small container with ten percent hypochlorite solution, then discarded in the waste container labeled “pipette tips”. Used tissues and masking tapes with labels were discarded in the biodegradable trash bin. The waste containers were transferred to a big drum at a university in a city in the Philippines. A third-party company collected the wastes gathered and transported them to Manila, Philippines.

Statistical treatments

The mean±SD of the physicochemical parameters and the concentration in mg/L of each of the heavy metals in the two sampling points with three replicates were computed. The means were compared with the standard range that was interpreted as either within allowable or beyond allowable levels of cadmium, copper, iron, lead, and zinc in the seawater.

Results

Physicochemical analysis: per Hydrogen (pH), temperature, and salinity

The physicochemical parameters including the pH, temperature, and salinity of the seawater samples collected from a barangay in a City in the Philippines during April 2023 were within the allowable levels (Table 1). The mean pH of the two sites was obtained and resulted in 8.45±0.38, which is within the standard range. The temperature was 30.00±1.10 °C, within the allowable levels. Additionally, the mean salinity obtained for the two sites was 30.55±0.27 ppt, which was within the allowable standard range.

Table 1. Physicochemical parameters of seawater in a Barangay in a City of the Philippines

Tested parameters	Mean±SD	Standard	Interpretation
pH	8.45±0.38	7.0 - 8.5	within allowable level
Temperature (°C)	30.00±1.10	26 - 30	within allowable level
Salinity (ppt)	30.55±0.27	> 30	within allowable level

Note: All standard physicochemical values were based on the Department of Environment and Natural Resources (DENR), Philippines Guidelines

Table 2. Concentrations and interpretations of heavy metals in seawater samples in a Barangay in Iloilo City

Kinds of heavy metals	Concs. of heavy metals (mg/L) Mean±SD	Standard (mg/L)	Interpretation
Cadmium	0.02±0.01	0.003	beyond allowable level
Copper	0.02±0.01	0.020	within allowable level
Iron	0.61±0.10	1.500	within allowable level
Lead	1.44±0.19	0.010	beyond allowable level
Zinc	0.02±0.01	0.050	within allowable level

Note: All standard physicochemical values were based on the Department of Environment and Natural Resources (DENR), Philippines Guidelines

Heavy metal analysis and their interpretations

The mean amounts and standard deviations of copper were 0.02±0.01, iron was 0.61±0.01, and zinc was 0.012±0.01mg/L. They were within the allowable standard range as of April 2023. The cadmium and lead were beyond the allowable standard range with the mean values of 0.02±0.02 and 1.44±0.19 mg/L (Table 2).

Discussion

Physicochemical analysis: per Hydrogen (pH), temperature, and salinity

The mean pH of the two sites was obtained and resulted in 8.45±0.38. In contrast to what Saalidong *et al.* (2022) asserted, this circumstance will not favor the event. They asserted that certain substances become toxic to aquatic life and that metals have a tendency to precipitate at extremely high pH values. Low pH, on the other hand, makes heavy metals in the water more toxic and toxicologically harmful because they become more soluble and bioavailable. It is generally known that when swallowed or applied to the skin, high water pH exposures can irritate the eyes, skin, and mucous membranes. Thus, in April 2023, it is important to measure the pH to get a sense of how the water is currently affected by heavy metals. The mean temperature of the two sites, 30.00+1.10°C, was within the permitted range. Copper's effect is not considerably affected by temperatures from 21 to 30 °C, but it became more harmful at high pH levels (Pascual *et al.*, 2022). According to the study's findings, the risk of copper is higher in colder climates than in warm ones. Algae and other marine organisms near coal-fired power plants did not encounter this condition as of April 2023 since the temperature was within acceptable ranges.

According to Zhou *et al.* (2019), salinity is often correlated with the toxicity of heavy metals, and when levels decrease, these impacts become more toxic. This is caused by the hazardous free metal ions' greater bioavailability under lower salinity conditions. No such condition existed in April 2023 since the seawater's salinity was within the standard range, 30.55+0.27 ppt.

Heavy metal analysis and their interpretations

Cadmium (Cd) levels in seawater must not exceed 0.003 mg/L. The mean cadmium content at points A and B in April 2023 was 0.02±0.01 mg/L. This could be taken to mean that the concentration is higher than what the DENR has allowed. The results of laboratory tests show that the amount of cadmium in saltwater pollution is almost entirely determined by the pollution produced by coal-fired power plant operations and that; this can be harmful to people's health. The human kidney may retain cadmium for up to 38 years, while the human liver can retain it for up to 19 years. It results in apoptosis, gene expression changes, membrane damage, and several metabolic and histological modifications (Delly *et al.*, 2021). Additionally, cadmium poisoning can affect the skeletal and cardiovascular systems, as well as impair eyesight and hearing, according to Kumar *et al.* (2019). Cadmium has significant teratogenic and mutagenic effects, but it also has harmful effects at low concentrations on both male and female reproductive, which can impair pregnancy. Exposure to Cd compounds is principally linked to an increased risk of pancreatic, lung, kidney, and prostate cancer. Breast, bladder, and urinary system malignancies have all been associated to Cd. The many mechanisms of Cd-induced carcinogenesis also involve interference with DNA repair mechanisms, lipid peroxidation increase, and oxidative stress with antioxidant enzyme suppression. Additionally, important metal ions, particularly redox-active ones, can be replaced by Cd^{2+} (Peana *et al.*, 2022).

Copper has a 0.02±0.01mg/L amount in salt water, which was within the allowable range according to the DENR. The body just requires a minimal amount of copper to function. Acute nausea may also develop from gastrointestinal tract irritation brought on by increasing exposure (Adams *et al.*, 2020). Nevertheless, the average copper concentration in the saltwater in a barangay of a city in the Philippines does not represent a serious risk to people who go swimming or fishing there during April 2023.

The DENR has set a limit of 1.5 mg/L for the amount of iron (Fe) in seawater. In April 2023, the average iron concentration was 0.61±0.10 mg/L, which was within the permitted range. This suggested that iron is unlikely to seriously poison the people living nearby. It is a hazardous heavy metal that can be obtained from man-made sources, such as coal-fired power plants, and is an essential heavy metal that is required for living organisms at specific doses (Scimino, 2021).

The DENR has determined that 0.01 mg/L of lead (Pb) is the maximum allowable content in seawater. The average lead concentration, according to the results, was 1.44±0.19 mg/L, which is above the permitted level during April 2023. As a result, lead poses a serious threat to both aquatic and terrestrial ecosystems. According to Munawer (2018), the high concentrations of lead in the saltwater near a coal-fired power plant are due to the easy release of lead into the environment. The high amount of lead pollutes the air and bodies of water in areas close to coal-fired power plants. Additionally, this is consistent with a study by (Delly *et al.*, 2021) in which lead and cadmium were found in seawater at high levels in the vicinity of a coal-fired power plant in a mining port near a residential area in the Morowali District.

The DENR has determined that 0.05 mg/L of zinc (Zn) is the permissible level. The mean zinc content in this study was 0.012±0.01mg/L. The outcome demonstrated that the zinc concentration was below the permitted range. It suggested that it could not have seriously hazardous effects on the residents living nearby. Toxic consequences on human health, such as gastrointestinal effects, immunological and lymph reticular effects, cardiovascular effects, carcinogenic effects, neurotoxicity, and toxicokinetics may arise if zinc discharge is not careful and is excessive (Sankhla *et al.*, 2019). Seawater close to coal-fired power stations contained high amounts of cadmium and lead had serious consequences for both the environment and human health last April 2023.

Effective pollution control techniques, frequent monitoring, and strict laws are required to handle this issue. By doing this, dangers that could result from heavy metal contamination can be avoided. The actions also can ensure the health of communities that depend on seafood as a primary food source and protect marine ecosystems.

The findings of this study were limited only to the seawater in a barangay of a city near the coal-fired power plant and were conducted in April 2023. Two sampling points and three replicates (three samples) per sampling point were analyzed.

Conclusion

This study provided additional information on the status of the concentration of heavy metals in seawater in the coastal area of the barangay near a coal-fired power plant. The seawater samples that were taken in April 2023 from a barangay in a City in the Philippines had all of their physicochemical parameters—pH, temperature, and salinity—within permissible bounds. The two sites' mean pH was measured, and the result was 8.45+0.38, which is within the acceptable range. The temperature was within permissible bounds at 30.00+1.10 degrees Celsius. Furthermore, the two locations' mean salinity of 30.55+0.27 ppt fell within the permitted standard range. The mean and standard deviations for zinc, iron, and copper were 0.012+0.01 mg/L, 0.61+0.01, and 0.02+0.02 mg/L, respectively. As of April 2023, they were inside the permitted standard range. Cadmium and lead levels, with mean values of 0.02+0.02 and 1.44+0.19 mg/L, respectively, exceeded the permitted standard range. This study established that the seawater near the coal-fired power plant needs continuous monitoring, as it contains cadmium and lead beyond allowable levels. However, the amount of copper, iron, and zinc might increase shortly due to the continuous operations of the coal fire-powered plant.

Recommendations

It is recommended to conduct a series of monitoring as to the level of heavy metals and increase sampling points as an extensive approach among the local government. Correspondingly, heavy metal concentrations in seawater sediments and edible marine organisms should be quantified. Other heavy metals such as mercury, arsenic, and manganese should be quantified also.

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