Tropical Aquatic and Soil Pollution

Research Article
Volume 1(2), 2021, 74-86
https://doi.org/10.53623/tasp.v1i2.20

Level and distribution of heavy metals in Miri River, Malaysia

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SUBMITTED: 22 July 2021; REVISED: 31 August 2021; ACCEPTED: 1 September 2021

ABSTRACT: Heavy metal pollution in water resources has become a serious and hazardous environmental problem all over the world because of its non-biodegradability, emanating from multiple sources, easy accumulation, and biological toxicity. This research was carried out to study the level and distribution of heavy metals at different sampling locations (upstream, midstream, and downstream), at different depths (0.5 m and 1.5 m from surface water level), and during low tide and high tide conditions in the Miri River of Miri City in Malaysia. The river water samples were collected and analyzed for Ca, Mg, Cu, Fe, Mn, Ni, Pb, and Zn by flame atomic absorption spectrophotometer. The concentration of Ca was found to be the highest in the Miri River, followed by Mg and Fe, and with traces of Cu, Mn, Ni, Pb, and Zn. An increase in the concentration of heavy metals, such as Cu, Mg, and Ni, was observed while flowing from upstream to downstream of the Miri River. Concentrations of heavy metals, such as Ca, Mg, Cu, and Zn, were clearly lower at 1.5 m depth than at 0.5 m depth. High tides in the river decrease the concentration of heavy metals, such as Ca, Cu, Mn, and Ni, compared to low tides. From this research, it gets clear that using the Miri River water for domestic and recreational purposes, washing, and fishing is detrimental to human health and the environment.

KEYWORDS: Heavy metals; Miri River; river water quality; depth; tides

1. Introduction

Water resources are an important natural resource for life on earth. Water is an irrefutable resource for domestic purposes, agriculture (e.g. irrigation, animal husbandry, etc.), various industries, energy production, and providing many ecosystem services (e.g. tourism, recreational activities, etc.). However, a variety of natural and anthropogenic activities degrade the quality of water resources and make them less usable [1,2]. Water pollution has become a serious concern, especially in developing countries, due to the direct discharge of wastewater into rivers and lakes without treatment [3–5]. At present, water quality is one of the key problems in the aquatic environment. Several point and non-point sources are contributing to the river's pollution [6]. The direct discharge of wastes and industrial by-products associated with toxic compounds, without proper treatment, into water resources represents an ongoing environmental problem due to their possible impact on the aquatic environment or river ecosystem, and a potential effect on public health [7]. The pollution of water resources directly affects the quality and quantity of water for irrigation and drinking water and increases the financial burden to treat or make it useable [6].

Trace metal pollution in water resources has become a serious and hazardous environmental problem all over the world because of its non-biodegradability, emanating from multiple sources, easy accumulation, and biological toxicity [8]. Trace metals are the dense metallic component, naturally occurring on the earth's crust, which are toxic to both aquatic and human life, due to their persistent and non-biodegradable characteristics with the ability to cause deleterious effects on living organisms [9]. Various solid and liquid wastes are generated as the by-products of manufacturing processes, which contain toxic chemicals such as chromium salts, sulfides and other substances, including heavy toxic trace metals [10]. Heavy metal pollution can occur through natural processes such as geological weathering, direct atmospheric deposition, etc., and anthropogenic activities such as agricultural, municipal, residential or industrial waste products [11]. A major source of energy for industries is coal combustion, which is one of the most important anthropogenic emission sources of trace elements and an important source of a number of metals [12].

Continuous accumulation of metals in water bodies can amplify the adverse effects on aquatic life and the water ecosystem. The human body requires some metals such as zinc, copper, iron, manganese, and cobalt for their development in very low concentrations. However, they may be toxic if ingested at higher concentrations [13]. Mostly, heavy metals enter the human body through ingestion (the consumption of contaminated drinking water or food) and may result in cardiovascular disorders, neuronal damage, renal injuries, and an increased risk of cancer and diabetes [14,15]. Trace metals can cause deoxyribonucleic acid (DNA) damage and lesions through the formation of reactive oxygen species in tissues [16]. Furthermore, irrigation with heavy metals-contaminated water degrades soil quality and has an impact on the overall agricultural system, including plant growth and yield, and thus the animals that consume the products [17–19].

Around 98% of the total water used in Malaysia is from rivers [20]. According to a report published by the Department of Energy, Malaysia in 2017, out of 477 rivers being monitored, about 11% of the rivers were found to be polluted and about 43% were slightly polluted. The Miri River is the major surface water source, flowing through Miri city. The Miri River starts inland, flows through Miri city, curves at Lutong town, and ends up in the South China Sea. The Miri River contributes 97% of the total water used, mainly for irrigation and domestic needs of the Miri city, all year round [21]. Hence, it is important to study the level and distribution of heavy metals in the water of Miri River. The objective of this research was to study the level of heavy metals in the Miri River, Sarawak, Malaysia and investigate the distribution of heavy metals at different sampling points in the river, depth of water in the river, and effects of tide on the concentration of the heavy metals.

2. Materials and Methods

2.1. Description of the Study Area

Miri is a coastal city in northeastern Sarawak, Malaysia, located near the border of Brunei, on the island of Borneo. The city is the capital of Miri District of the Miri Division and the second largest city in Sarawak. The population of the city is increasing every year, which has led to the city's emerging as an industrial district, and many factories are built in the city, along the river. The establishment of these factories often entailed significant environmental footprints, due to the direct disposal of biological and chemical wastes of the factories in the Miri River. One of the major pollutants in the river is heavy metals. The heavy metals that pervaded into the river would pollute the river, which would result in poor water quality conditions.

2.2. Water Sampling

Water sampling locations were determined at the upstream, midstream (about 7.5 km downstream from the upstream locations), and downstream (about 7.4 km downstream from the midstream locations) of the Miri River, and samples were collected from 2 points in each location by the grab method. The exact location and description of the sampling points are presented in Table 1. Water samples were collected at 0.5 m and 1.5 m depth from the surface water level of each sampling point, to check the variation in concentrations at different depths. Also, the water samples were collected during low tide and high tide periods, to understand the effect of tides on the variation of concentrations.

Table 1. Location (coordinates) and description of the sampling points.

	Coordinate of point 1	**Coordinate of point 2**	**Description**
Upstream	4.412236,114.019651	4.411019,114.020787	Heavy industrial and residential area
Midstream	4.468002,114.009296	4.467889,114.012893	Commercial area
Downstream	4.400356,113.986662	4.399246,113.986053	Fishery and industrial area

2.3. Preparation and Analysis of Samples

The collected water samples were carried to the laboratory, acidified with 20% nitric acid, digested, filtered, and analyzed for eight different heavy metals, namely lead (Pb), iron (Fe), manganese (Mn), copper (Cu), nickel (Ni), magnesium (Mg), calcium (Ca), and zinc (Zn). The samples were analyzed by using a Flame Atomic Absorption Spectrophotometer (AA400, Perkin Elmer), following Standard Methods for the Examination of Water and Wastewater [22]. Various concentrations of standard solutions of the same heavy metal were prepared by dilution, and were used to calibrate and analyze the river water samples. A different wavelength of source lamp and a different type of lamp were used for the analysis of each heavy metal.

3. Results and Discussion

The results or concentration of each heavy metal at different sampling points (upstream [U/S], midstream [M/S], and downstream [D/S]) of the Miri river, at different depths (0.5 m and 1.5 m), and the effect of tides (low tide [LT] and high tide [HT]) on the variation of concentration are presented and discussed individually for each metal.

3.1. Calcium (Ca)

Figure 1 depicts Ca concentrations upstream, midstream, and downstream of the Miri River, at 0.5 m and 1.5 m depths, as well as variation in concentrations at low and high tide conditions. The Ca concentration in the Miri River was found to be widely varied in the range of 113 to 1,545 mg/L. The Ca concentration was found to be fluctuating in the upstream, midstream, and downstream sampling locations and different sampling depths. In general, the concentration of Ca in rivers is low, but it can be high due to flow through the lime areas or dumping of waste containing Ca. Human activities (domestic and industrial) in the vicinity of the Miri River might have contributed to the higher concentration of Ca. Sources of Ca can be some industrial waste waters from processing, in which the acids are neutralized by lime or limestone [23]. Though there is no health-based value for the concentration of Ca in the WHO guidelines for drinking water, the taste threshold for the Ca ion is in the range of 100-300 mg/L [13]. Also, there remains the possibility of scale deposition in the treatment works, distribution system and pipeworks of the water containing high Ca concentrations. There is no limit provided in the guidelines for Ca concentration in the National Water Quality Standards (NWQS) for Malaysia [24], but the water hardness should be paid attention to before using the river water for any type of use.

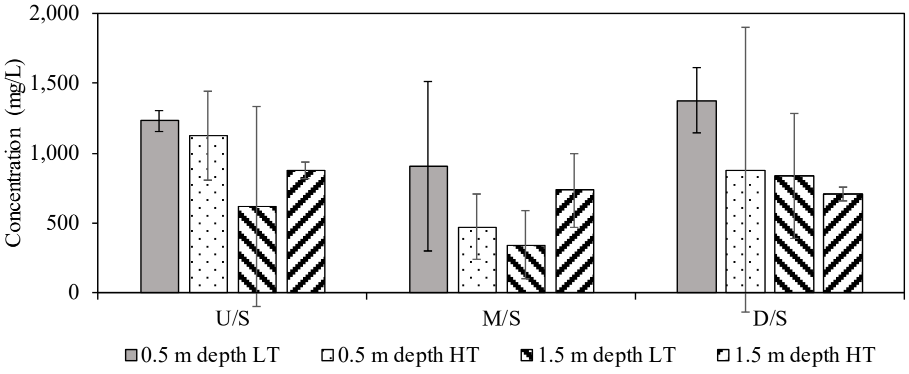

Figure 1. Calcium (Ca) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. (*n* = 2).

3.2. Magnesium (Mg)

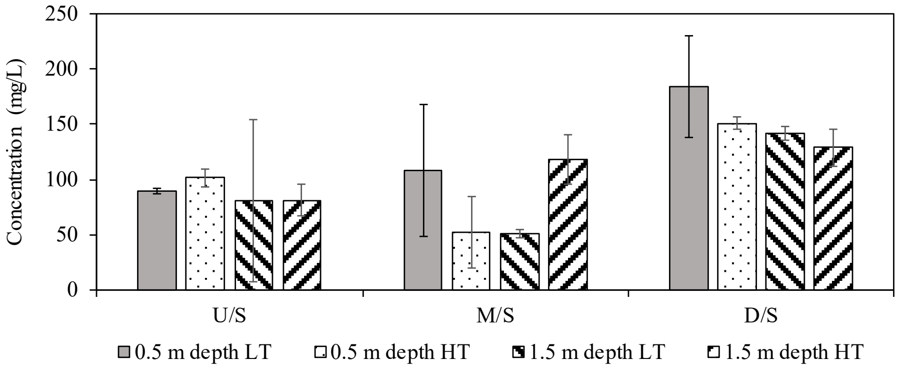

Figure 2. Magnesium (Mg) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. (*n* = 2).

Figure 2 depicts the concentration of Mg upstream, midstream, and downstream of the Miri River, at 0.5 m and 1.5 m depths, as well as the variation in concentration at low and high tide conditions.The Mg concentration in the Miri River ranged from 29 to 217 mg/L. There is no health-based value for Mg concentration in the WHO guidelines for drinking water, but it can increase the hardness of water, limiting the usability of the water. The Mg concentration was found to be fluctuating in the upstream, midstream, and downstream sampling locations and different sampling depths. However, the Mg concentration was found to be increasing from upstream to downstream, which might be due to direct discharge of wastes into the river downstream. The Mg concentration was lower during high tide than during low tide. It might be due to the dilution of Mg in a higher volume of water, during high tide conditions.

3.3. Copper (Cu)

Cu is an important trace element (recommended daily allowance of 2.0-3.0 mg/day for humans) for several enzymes involved in antioxidant response, membrane and DNA integrity, and adenosine triphosphate production [25,26]. Cu accumulation in bacteria can cause protein damage and cell injury [26,27], and high levels of Cu exposure can cause toxicity and the development of copper resistance in a variety of pathogenic bacteria [28,29]. Concentration of Cu at upstream, midstream, and downstream of the Miri River, at 0.5 m depth and 1.5 m depth, and variation in concentration at low tide and high tide conditions are presented in Figure 3. The Cu concentration in the Miri River ranged from 0.03 to 0.13 mg/L. Naturally occurring concentrations of Cu in the freshwaster systems have been reported to be from 0.20 to 30 μg/L [30]. It was observed that the Cu concentrations increased from upstream to downstream and the concentration was lower during high tide than that during low tide. Human activities and the establishment of industries near the Miri River might have contributed to the anthropogenic sources of the increase in concentration of Cu. According to the NWQS for Malaysia [24], the Miri River water generally did not conform to the established standard for drinking water and recreations involving body contact. The water might also impact the fishery as the water was not suitable for sensitive and very sensitive aquatic species. The water could be used for irrigation as well as deemed acceptable for common aquatic species.

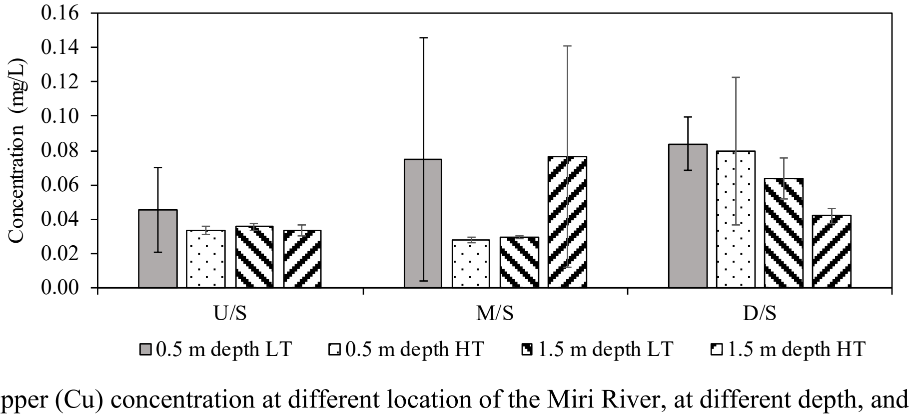

Figure 3. Copper (Cu) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. (*n* = 2).

3.4. Iron (Fe)

Fe is one of the most abundant metals in the Earth's crust and an essential element in human nutrition. No health-based value has been provided for the concentration of Fe in the WHO guidelines for drinking water [13], but it is preferred below 0.3 mg/L, as the taste of Fe is unnoticeable below that value. Higher concentrations of Fe affect the taste of drinking water and stain clothes during laundry. Concentration of Fe at upstream, midstream, and downstream of the Miri River, at 0.5 m depth and 1.5 m depth, and variation in concentration at low tide and high tide conditions are presented in Figure 4. The Fe concentration in the Miri River ranged from 1.45 to 7.63 mg/L. The Fe concentration in the upstream (> 5 mg/L) was significantly higher than that in the midstream and downstream. According to the NWQS for Malaysia [24], the Miri River water upstream was not suitable for irrigation purposes. Higher concentrations of Fe at the upstream and lower concentrations of Fe at the midstream and downstream might be due to settling of the Fe-sediments in the river-bed during flow or the discharge of Fe wastes near the upstream sampling points.

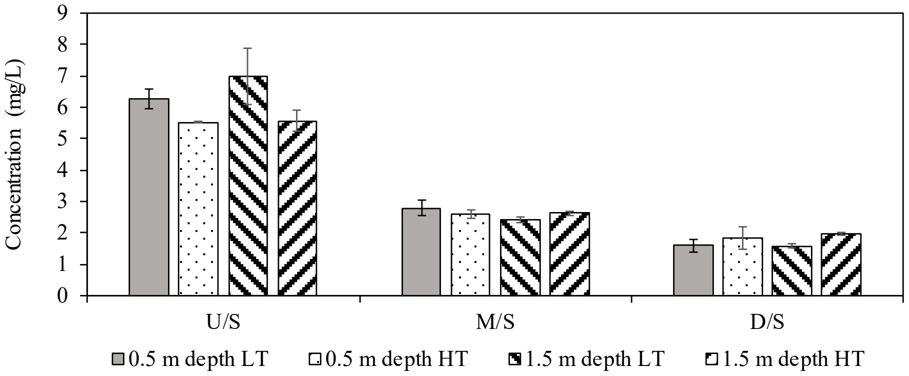

Figure 4. Iron (Fe) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. ($n = 2$).

3.5. Manganese (Mn)

Mn is also an abundant metal in the Earth's crust and is commonly found with Fe. The concentration of Mn in naturally occurring surface waters ranged from 1 to 200 µg/L. A health based value of 0.4 mg/L of Mn concentration has been considered in the WHO guidelines for drinking water [13]. If the concentration of Mn is below 0.1 mg/L, then both guidelines for organoleptic properties and health guidelines can be met [31]. Concentration of Mn at upstream, midstream, and downstream of the Miri River, at 0.5 m depth and 1.5 m depth, and variation in concentration at low tide and high tide conditions are presented in Figure 5. The Mn concentration in the Miri River ranged from 0.03 to 0.48 mg/L. The concentration of Mn at the upstream and midstream was less than 0.11 mg/L (at both sampling depths and tidal conditions), whereas the concentration was higher downstream. It might be due to the disposal of waste containing Mn or Mn compounds in the downstream. According to NWQS for Malaysia [24], the Miri River water upstream and downstream can be used for drinking and recreational purposes after proper treatment. From the point of view of concentration of Mn, however, the Miri River water downstream was not fit for drinking and recreational purposes as well as it could cause adverse impacts on the fishing industry, but it can be used for irrigation. The Miri River water upstream and midstream can also be used for fishing of sensitive and common aquatic species. This might be the reason for the establishment of many fisheries downstream of the Miri River.

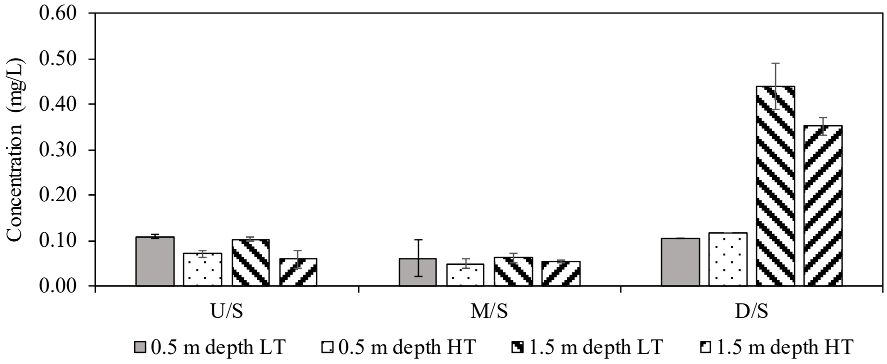

Figure 5. Manganese (Mn) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. (*n* = 2).

3.6. Nickel (Ni)

The maximum recommended value of Ni based on human health criteria for drinking water is 0.07 mg/L. Higher concentrations of Ni in water cause several health risks to plants and animals [13]. Figure 6 depicts the concentration of Ni upstream, midstream, and downstream of the Miri River, at 0.5 m and 1.5 m depths, as well as the variation in concentration at low and high tide conditions.The Ni concentration in the Miri River ranged from 0.12 to 0.26 mg/L. The concentration of Ni in the Miri River water was higher than the recommended value. It might be due to the release of Ni and Ni compounds from natural or industrial Ni wastes into the river bank. Ni concentrations increased downstream more than upstream.It might be due to the disposal of industrial wastes containing Ni in the downstream, without proper treatment. In general, the concentration of Ni was lower at higher depths (i.e., 1.5 m) than at lower depths (i.e., 0.5 m), and also at higher tides than at lower tides.It might be due to the dilution of Ni in the higher volume of river water during high tide conditions. According to NWQS for Malaysia [24], the Miri River water was found to be unfit for drinking and recreational activities involving body contact, from the aspect of Ni contamination. However, the concentration of Ni in water at the upstream and midstream of the Miri River was less than 0.2 mg/L, which indicates that the river water at the upstream and downstream can be used for irrigation purposes.

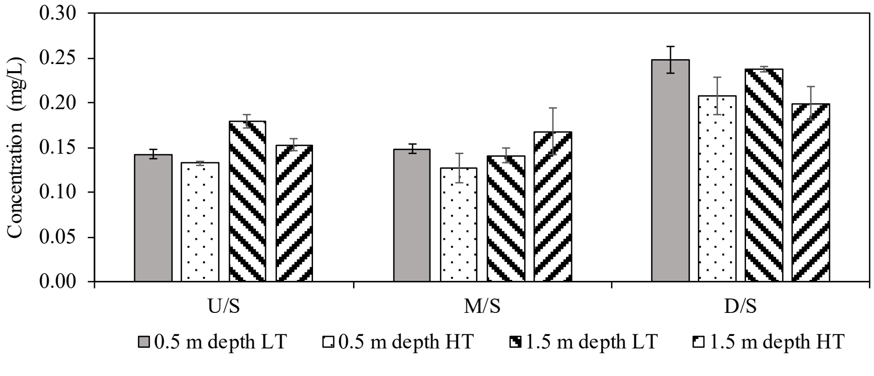

Figure 6. Nickel (Ni) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. (*n* = 2).

3.7. Lead (Pb)

Pb is one of the commonest heavy metals. Pb has been recognized and highlighted as one of the most dangerous environmental poisons [32]. The most recent guideline value for Pb in drinking water from the WHO is 10 μg/L but it is no longer a health-based value and has been designated provisional [13], due to an understanding of the harmful impacts of Pb on health. Figure 7 depicts the concentration of Pb upstream, midstream, and downstream of the Miri River, at 0.5 m and 1.5 m depths, as well as the variation in concentration at low and high tide conditions.The Pb concentration in the Miri River fluctuated from not detected to 0.35 mg/L. The Pb concentration in the normal river ranges from 0.003 to 0.03 mg/L. This indicates that the Miri River is polluted in the aspect of Pb concentration too. Though any particular trend of variation in Pb concentration at the upstream, midstream, and downstream sampling locations, depths, and tidal conditions was not obtained, it can be observed that the Miri River water was not suitable for drinking, and recreational activities involving body contact. According to NWQS for Malaysia [24], the Miri River water could also impact the fisheries or aquatic species of sensitive nature, and is therefore recommended for irrigation purposes only. Pb usually gets into water bodies due to leaching from lead-containing service lines, lead solder, and brass fittings, particularly in corrosive waters. There might be similar industries along the Miri River discharging their waste without proper treatment.

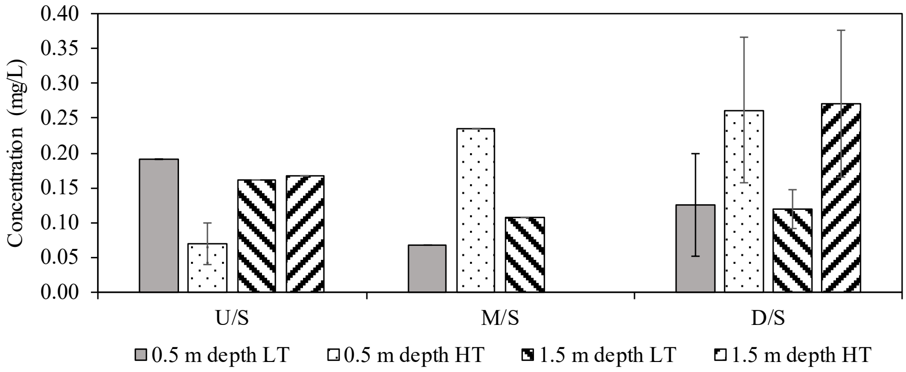

Figure 7. Lead (Pb) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. (*n* = 2).

3.8. Zinc (Zn)

Zn is an essential element required by humans for optimum growth and development, but the presence of a high amount of Zn in water may cause a bitter taste and opalescence in water. Though there is no health based value for Zn concentration in the WHO guidelines for drinking water, the preferred limit is 3 mg/L [13]. Concentration of Zn at the upstream, midstream, and downstream of the Miri River, at 0.5 m depth and 1.5 m depth, and variation in concentration at low tide and high tide conditions are presented in Figure 8. The Zn concentration in the Miri River ranged from 0.04 to 0.18 mg/L. This indicates that the Miri River water is fairly acceptable for every use in terms of Zn concentration [24]. However, no particular trend or variation in Zn concentration at the upstream, midstream, and downstream sampling locations, depths, and tidal conditions was obtained.

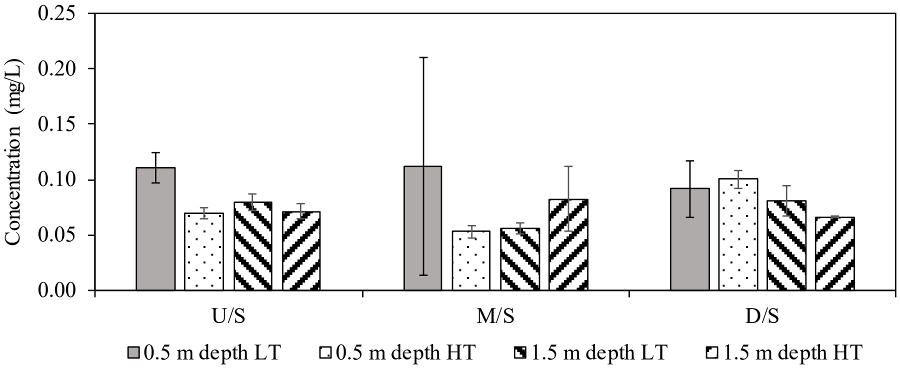

Figure 8. Zinc (Zn) concentration at different location of the Miri River, at different depth, and variation on concentration due to tidal effect. The plots present mean values of 2 sampling points at each location and the error bars correspond to S.D. (*n* = 2).

3.9. Heavy Metals in the Miri River

Levels and distributions in concentration of eight heavy metals, namely Ca, Mg, Cu, Fe, Mn, Ni, Pb, and Zn, were studied at the upstream, midstream, and downstream sampling locations, at 0.5 m depth and 1.5 m depth, and at high tide and low tide conditions in the Miri River. Concentrations of Ca were found to be the highest in the Miri River, followed by Mg and Fe. Other metal concentrations (Cu, Mn, Ni, Pb, and Zn) were present in traces, but few of them exceeded the permissible limits of drinking water, and some of them could not even be used for recreational or fishing purposes (discussed individually in the previous section).Previous studies by Wogu and Okaka in the Warri River in Nigeria [33], which receives industrial, agricultural, and urban sewage, showed that the concentrations of Cd, Cr, Mn, and Ni were higher than the standard levels, and the water was harmful to public health and hygiene. The results showed an increase in the concentration of heavy metals from upstream to downstream, especially for Cu, Mg, and Ni. The results of the analysis of water and sediment samples upstream and downstream of the Tembi River in Iran [34] also revealed that the mean concentration of several heavy metals in the downstream river water was significantly higher than that in the upstream river water. As the water flows downstream of the river, the dissolved heavy metals and sediments upstream are also transported downstream. Water physicochemical properties may also influence heavy metal precipitation in sediments [35], contributing to an increase in heavy metal concentrations downstream.In addition to this, human activities and direct disposal of industrial wastes in the vicinity of the Miri River in the midstream and downstream might have increased the concentrations of heavy metals downstream of the Miri River.

Anomalies in the variation of the concentration of some heavy metals upstream and downstream existed. For example, the concentration of Fe was higher in the upstream than in the downstream. It could be due to the sampling locations in the upstream, being near to the discharge points of domestic and industrial wastes. A similar phenomenon was observed in a study on the Yamuna River in India [36]. Because of the presence of several ironworks or related industries in the downstream area, the Fe concentration increased many folds. of heavy metals, such as Ca, Mg, Cu, and Zn, were clearly lower at 1.5 m depth than at 0.5 m depth, while fluctuating results were observed for other metals. In general, the concentration of heavy metals increased with depth of sampling in a study of the depth variability of heavy metal concentrations in the water of Firiza-Strimtori Lake, NW of Romania [37]. It might be due to the emanation of the metal ions into the water as an endogenic source of contamination based on the alteration in the re-dox potential of sediments. Anomalies in the results of other metals might be due to the variation in pH, redox potential, and river flow characteristics (e.g., flow rate) [38]. Concentrations of heavy metals, such as Ca, Cu, Mn, and Ni, were observed to be lower in high tide conditions than in low tide conditions, whereas no clear trend was obtained in remaining metals. The decrease in concentration at high tide conditions can be directly linked to the dilution of the metals in huge volumes of water during high tide, in comparison to the water volume during low tide. Tidal currents can affect the shifting of pollutants along the estuary, moving them upstream during high tide and downstream during low tide [38], which could be one reason for the anomalies in the variation of heavy metal concentrations upstream and downstream.

Apart from these parameters, the level and distribution of heavy metals depend on the surrounding environmental factors and several physico-chemical characteristics [39,40]. Fine-grained sediments have higher surface area for adsorption and ionic attraction, so the concentration of heavy metals, generally, increases with the smaller particle size of sediments [41,42]. Moreover, the transportation of sediments, along with the heavy metals, downstream of the river was also higher with the fine-grained sediments [42]. The pH, dissolved oxygen, temperature, and flow rate of the river water also affect the release of heavy metals from sediments [43]. Higher hydrogen ion concentration at lower pH tends to occupy more adsorption sites in sediments, resulting in easier precipitation of soluble and carbonate-bound heavy metals. Heavy metals can exist in the combined form with organic matters, which will oxidize with higher dissolved oxygen in the water, and release heavy metals from organic matters in water. Higher temperatures can accelerate the release of heavy metals in water from water-soluble fractions, carbonate fractions, and exchangeable fractions of the sediments. With the higher flow rate, there remains the possibility of higher dissolved oxygen and the physical disturbance in the sediments, which can promote the oxidation/reaction to release the heavy metals in water. However, these parameters were not considered in this study.

4. Conclusion

The level of heavy metals at different sampling locations (upstream, midstream, and downstream), at different depths (0.5 m and 1.5 m from surface water level), and at low tide and high tide conditions of the Miri River were studied. The concentration of Ca was found to be the highest in the Miri River, followed by Mg and Fe. Other metal concentrations (Cu, Mn, Ni, Pb, and Zn) were present in traces, but few of them exceeded the permissible limits of drinking water, and some of them could not even be used for recreational purposes involving body contact, such as fishing.in the concentration of heavy metals, such as Cu, Mg, and Ni, was observed from upstream to downstream of the Miri River, whereas some anomalies existed. Concentrations of heavy metals, such as Ca, Mg, Cu, and Zn, were clearly lower at 1.5 m depth than at 0.5 m depth, while results fluctuated for other metals. Concentrations of heavy metals, such as Ca, Cu, Mn, and Ni, were observed to be lower during high tide conditions than during low tide conditions, whereas no clear trend was obtained in remaining metals. In general, the Miri River water was found to be polluted with heavy metals, at different concentration levels. According to the NWQS for Malaysia, Miri River water can be considered suitable for irrigation purposes only. It is recommended to conduct an integrated and comprehensive study of the heavy metal concentrations in the Miri River, considering all the affecting parameters, to better understand the distribution of heavy metals and sources of contamination.

Acknowledgments

The authors thank Curtin University Malaysia for facilitating this study. Collaboration from Yamanashi University is highly appreciated.

Competing Interest

All authors declare no competing interests.

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