



Assessment of metal concentrations from recreational rivers in a tropical region (Jengka, Malaysia)

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Abstract

A recreational river may be exposed to some extent of metal pollution. Two rivers, namely Sungai Weh and Sungai Jempul, were selected for this study, as both areas offer recreational activities for residents in Jengka, Pahang, Malaysia. This study examines the concentration of selected metals (Fe, Mn, and Pb), elucidates the possible sources, evaluates the toxicity loads, and estimates the potential health risk. Metal concentrations were analysed using inductively coupled plasma optical emission spectrometry (ICP-OES). The concentrations were found in the sequence of Fe > Pb > Mn. Hierarchical cluster analysis (HCA) suggested that the metals' origins included both anthropogenic activities and natural sources. About 16.66% of Pb should be removed from the river water to ensure safety, based on the heavy metal toxicity load calculation (HMTL). Children are more vulnerable to non-cancer and cancer risks than adults. The findings indicate that comprehensive monitoring of water quality parameters and thorough exposure assessment should be performed.

Keywords Metal · Recreational river · Surface water · Water quality

Introduction

River water is a source of drinking water, agricultural, industrial and recreational activities. Nevertheless, the river water quality may deteriorate due to increases in human population and anthropogenic activities. The discharge of industrial by-products, untreated domestic waste and agricultural waste (Ekka et al. 2020; Ling et al. 2017; Islam et al. 2013; Rahman et al. 2013) and lack of awareness among the citizens (Suratman et al. 2015) are pollution sources of river water. Metals are an environmental pollution source that can weaken the river water quality and consequently disturb the river ecosystem. Exposure to metals in river water can lead to harmful effects on human health. Metals such as iron (Fe) and manganese (Mn) are required for the human

body in a specific amount. However, if the concentration is high, they could cause chronic health problems such as kidney malfunction, anaemia and liver problems (Said et al. 2011; Juen et al. 2014). Moreover, previous research on fish samples from recreational rivers indicated that Fe, Mn and Pb are traceable (Harith et al. 2021). Therefore, regular monitoring of rivers is vital to assess the water quality for numerous benefits (Sun et al. 2019). Additionally, the Sustainable Development Goals (SDGs) have emphasized the importance of clean water access (UN 2015).

Sungai Jempul and Sungai Weh are located in Jengka, Pahang, Malaysia. The rivers are located in a suburban region that is well-known for its palm oil plantations. The typical daily temperatures are between 24 °C and 35 °C, with an annual rainfall of 2000 mm. Both rivers provide water resources and local fish supply and flow through reserved forests, agricultural areas, and development (Harith et al. 2021). Apart from supplying water to local citizens, both rivers are recognized as recreational areas. These locations are the main attractive ecotourism area for residents. Several studies indicated the recreational rivers have the tendency to be polluted (Jang 2016; Massoud 2012; Holtcamp 2012). Therefore, it is vital to evaluate the level of metal pollution from Sungai Jempul and Sungai Weh.

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Numerous approaches, such as the ecological risk index (ERI) and the hazard index (HI), are used to analyse and compare the quality of waterways. Typical evaluation techniques are incapable of predicting the percentage of harmful heavy metals that should be removed from water bodies to limit the health risks (Mukanyandwi et al. 2019; Proshad et al. 2020). Saha and Paul (2019) created a unique evaluation method named heavy metal toxicity load (HMTL), which can accurately determine the level of toxic heavy metals in the water that may pose a health risk to humans. This assessment approach can also be used to calculate the required proportion of heavy metals to be eliminated from water bodies (Saha and Paul 2019). Several studies around the world applied this approach, such as in China (Huang et al. 2021), Nigeria (Ayejoto et al. 2021), Bangladesh (Proshad et al. 2020), and India (Saha and Paul 2019). Since this approach has been recently developed, there is no extensive study in the existing literature that uses it to examine the recreational river water quality, determine toxicity levels and infer potential human health hazards especially in tropical region. Additionally, there is no reported study on metal concentrations from two selected recreational rivers (Sungai Jempul and Sungai Weh) in Jengka, Pahang, Malaysia. Therefore, metal concentrations from these recreational rivers were determined and the sources were identified. This new method aims to evaluate the metal toxicity and potential health risks to the local population.

Materials and methods

Sample collection and analysis

About 30 samples were collected from Sungai Jempul (3°45'10" N, 102°39'5" E) and Sungai Weh (3°39'9" N, 102°41'43" E) in Jengka, Pahang, Malaysia (Fig. 1). Both rivers flow through reserved forests, agricultural areas, and development activities. These rivers originate from different water catchment areas, however, connected as tributaries of the Jengka river. The distance between these two rivers was about 50 km.

One sampling event was carried out during hot days. Wet days were not considered for sampling due to the possible dilution effect of pollutants. The samples were collected from the top surface of the water (< 10 cm depth) in the middle area of the river to avoid any misrepresentation of data using the grab sampling technique. Samples were taken from upstream to downstream at an interval of 500 m between 5 sampling points for each river. The water samples were collected directly into 5% HNO₃ pre-acid-washed 250 ml polyethylene (PE) bottles with polyethylene caps. The water samples were acidified to pH < 2, stored in an icebox before being brought to the laboratory,

and kept at 4 °C prior to further analysis. All samples were filtered through a 0.45 µm pore size membrane filter before analysis. The metal (Fe, Mn and Pb) concentrations in the filtered samples were then analysed using inductively coupled plasma optical emission spectrometry (ICP-OES). Each sample was spiked with the target analytes of interest in the spike recovery test. The recovery rates ranged from 85 to 110%. The detection limits of Fe, Mn and Pb were 0.2, 0.01 and 0.005 mg/l, respectively.

Heavy metal toxicity load (HMTL)

Heavy metal toxic load (HMTL) is a measurement of the quantity of heavy metal in water that may be harmful to humans. It indicates how much treatment is needed to make the water appropriate for human usage (Huang et al. 2021). This method also aids in the documentation of an effective treatment and management strategy. HMTL is calculated by multiplying the detected heavy metal concentration by the hazard intensity, as shown in Eq. 1:

$$\text{HMTL} = \sum_{i=1}^n C \times \text{HIS} \quad (1)$$

where C is the concentration of heavy metals, n is the number of heavy metals, and HIS is the hazard intensity score determined from the ATSDR (ATSDR, 2019). HIS is determined by the frequency of toxic metals as a dangerous substance on ATSDR's National Priorities List (NPL) sites, the toxicity level of the metals, and the possibility of human interaction. The HIS value is 797 points for Mn and 1531 points for Pb.

Metal evaluation index (MEI)

The metal evaluation index (MEI) measures the water's overall quality in terms of heavy metals (Edet and Offiong 2002) and was calculated using Eq. 2:

$$\text{MEI} = \text{Mc} / \text{Mmac} \quad (2)$$

where Mc is the metal concentration and Mmac is the maximum allowable concentration of the metal. The maximum allowable concentrations for Fe, Mn, and Pb are 1.00, 0.10 and 0.05 mg/l, respectively (NDWQS 2019). According to WHO (2011), the maximum allowable concentration for Mn is 0.40 mg/l and that for Pb is 0.01 mg/l. The MEI can be categorised as low (less than 10), medium (between 10 and 20), or high (more than 20).

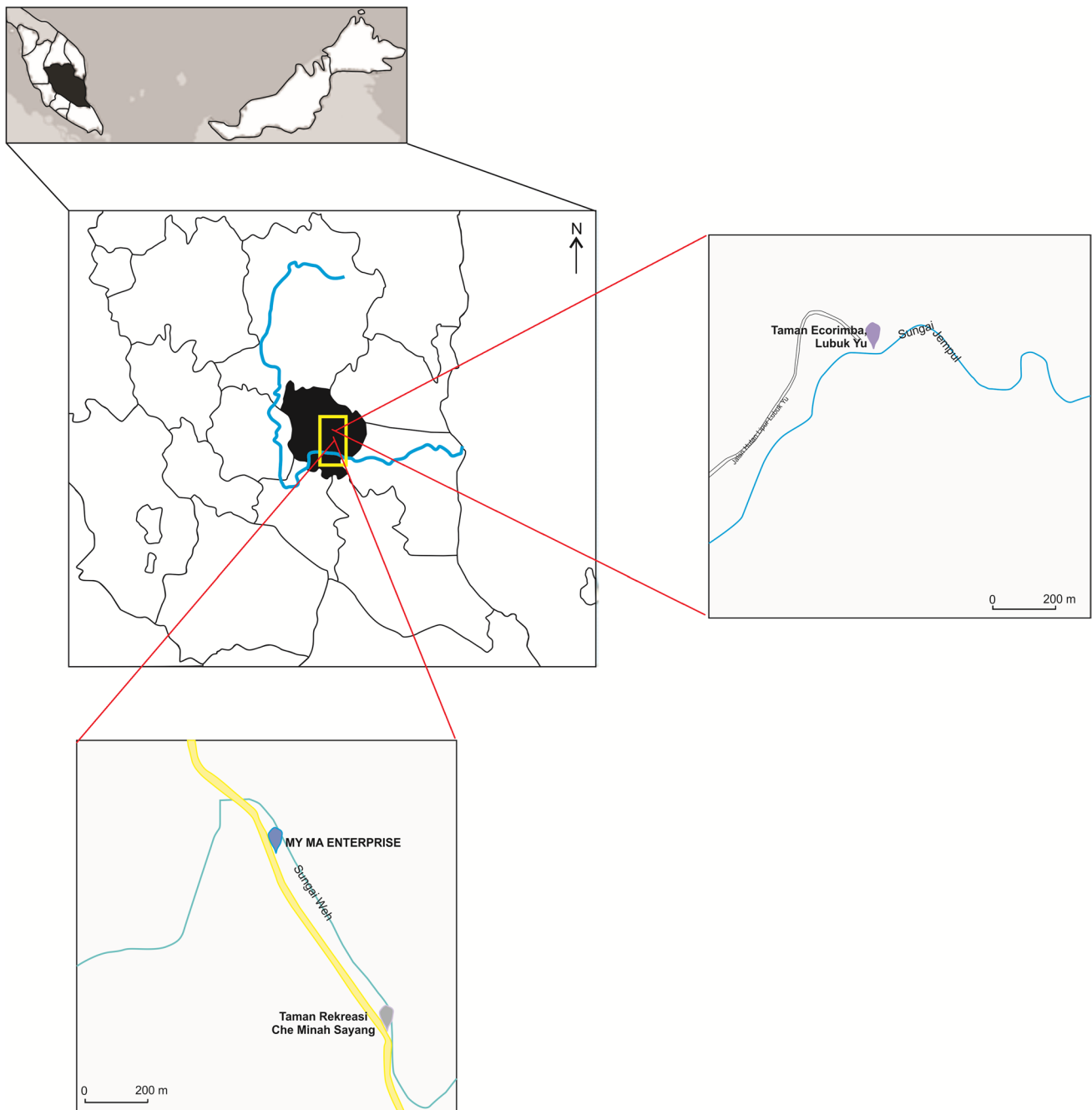


Fig. 1 Sampling locations in Sungai Jempul and Sungai Weh, Jengka, Pahang, Malaysia

Human health risk estimation

Heavy metals in surface water enter the human body via ingestion and dermal contact. Empirical models can predict whether oral consumption or dermal contact will have non-carcinogenic or carcinogenic health effects. The average daily dose (ADD) is a tool for estimating the daily impact of heavy metals in contaminated water on human health and was calculated using Eqs. 3 and 4. The hazard quotient (HQ) was

used to calculate the assessment of health risks in relation to non-carcinogenic effects as based on ADD (Eq. 5). The hazard index (HI) was computed by adding the individual HQs as expressed in Eq. 6 (Sulaiman et al. 2020).

$$ADD_{\text{oral}} = (C_{\text{water}} \times IR \times ED \times EF) / (BW \times AT) \quad (3)$$

$$ADD_{\text{dermal}} = (C_{\text{water}} \times SA \times AF \times ABF \times ED \times EF) / (BW \times AT) \quad (4)$$

$$HQ = ADD/RfD \quad (5)$$

$$HI = \sum HQ \quad (6)$$

The incremental risk of an individual acquiring cancer throughout a lifetime as a result of exposure to a potential carcinogen can be estimated as the carcinogenic risk (CR) for oral and skin contact of contaminated water (Proshad et al. 2020). The following formulae were used to determine the cancer risk in this study:

$$CR_{\text{oral}} = ADD_{\text{oral}} \times SF \quad (7)$$

$$CR_{\text{dermal}} = ADD_{\text{dermal}} \times SF \quad (8)$$

$$TCR = CR_{\text{oral}} + CR_{\text{dermal}} \quad (9)$$

The USEPA (2011) defines an acceptable range of cancer risk between 1.0×10^{-6} to 1.0×10^{-4} . Supplementary Table S1 contains the values of the variables (ABF, AF, AT, BW, IR, ED, EF, SA, RfD, SF) used in the calculation of human health risk exposure to water (Malaysian Adult Nutrition Survey 2009; USDOE 2011; USEPA 2002, 2011).

Statistical analysis

The results were analysed using IBM Statistical Package for Social Sciences (SPSS Version 21). One-way analysis of variance (ANOVA) was used to determine any significant differences between the parameters at a 99% confidence level. Pearson correlation was utilised to verify the relationship between the metals as the data gained were normally distributed. Hierarchical cluster analysis (HCA) was applied to categorise the metal according to similar potential pollution sources.

Results and discussion

Metal concentrations

Table 1 shows the descriptive statistics of metal concentrations (Fe, Mn, Pb) in river water samples from Sungai Jempul and Sungai Weh. The highest mean of Fe concentration was 0.48 mg/l for Sungai Jempul and 0.31 mg/l for Sungai Weh, followed by Pb concentration means of 0.17 mg/l (Sungai Jempul) and 0.19 mg/l (Sungai Weh). The lowest mean concentration was Mn with 0.07 mg/l for Sungai Jempul and 0.02 mg/l for Sungai Weh. Note that Fe concentration was the highest, as this element is the most abundant in the Earth's crust (Lim et al. 2013). Fe and Mn concentrations were both within the maximum acceptable concentration set by WHO (2011) and NDWQS (2019). Pb concentrations, however, showed values higher than the limit. Significant differences ($p < 0.001$) between metals in these two rivers were observed based on ANOVA results, except for Pb. This finding hints that Pb could come from different sources.

Heavy metal toxicity loads in water

Heavy metal toxicity loads (HMTL) are used to assess the level of toxic metals in water bodies and determine the concentration of metals that should be removed for the water to be safe for human use. Kumar et al. (2019) stated that the HMTL index provides the toxicity level of a pollutants in water that results in a non-carcinogenic risk and aids in the treatment and management. The HMTL was determined for Mn and Pb according to the ATSDR's substance priority list (ATSDR 2019). The permissible concentrations (mg/l) are 0.3 and 0.65 for Mn and Pb, respectively (ATSDR 2019). The HMTL for Sungai Jempul was 55.79 for Mn and 260.27 for Pb, whereas the values for Sungai Weh were 15.94 for Mn and 290.89 for Pb (Table 2). The HMTL in this study is lower than the maximum toxicity load, indicating a low level of harmful

Table 1 Descriptive statistics for Fe, Mn, and Pb concentration (mg/l)

		Min	Max	Mean	SD	Variance	p value	MAC*	
								WHO (2011)	NDWQS (2019)
Sg. Jempul (n=15)	Fe	0.45	0.51	0.48	0.02	5.10	0.000	n/a	1.00
	Mn	0.06	0.08	0.07	0.01	6.07	0.000	0.40	0.10
	Pb	0.16	0.18	0.17	0.007	5.38	0.104	0.01	0.05
Sg. Weh (n=15)	Fe	0.25	0.40	0.31	0.06	5.49	0.000	n/a	1.00
	Mn	0.01	0.03	0.02	0.008	6.20	0.000	0.40	0.10
	Pb	0.16	0.22	0.19	0.02	5.78	0.110	0.01	0.05

n/a Not available, *Maximum acceptable concentration (MAC), World Health Organization (WHO), National Drinking Water Quality Standard (NDWQS), Bold values indicate greater than MAC

Table 2 Heavy metal toxicity load (HMTL, mg/l) of the river water based on the relative level of heavy metals

	Mn	Pb	HMTL
Sg. Jempul	55.79	260.27	316.06
Sg. Weh	15.94	290.89	306.83
Total	71.73	551.16	622.89
Hazard intensity score (HIS) ^a	797	1531	
Permissible toxicity load (mg/l)	239.1	459.3	
% Removal of metal to reduce pollution load	PTL	16.66	

^aATSDR 2019, PTL permissible toxicity load

metal pollution in water. Continuous water contamination, nonetheless, has the potential to raise the HMTL even higher. The total HMTL suggests 16.66% of Pb in river water could be removed from both rivers.

The HMTL is a new approach to evaluate heavy metal toxicity load that can accurately determine the level of heavy metals in the water that may pose a health risk (Saha and Paul 2019). Several studies in other parts of the world adapted this method (Huang et al. 2021; Ayejoto et al. 2021; Proshad et al. 2020). However, there was still limited study using this approach in the tropical region to evaluate the recreational river. This study adopted this method in assessing the metal toxicity load in river water, and the same approach can be applied to other rivers in the same region.

Metal evaluation index

The metal evaluation index (MEI) is a tool used to measure the water quality that focuses on heavy metals. It gives total water quality in terms of heavy metals. Table 3 presents the MEI values computed for Fe, Mn, and Pb. In this study, MEI was determined based on WHO (2011) and NDWQS (2019) maximum allowable concentration. According to WHO, the MEI value was classified as low for Mn but medium for Pb. However, if based on NDWQS, the MEI for Fe, Mn, and Pb was considered low. In general, heavy metal pollution levels in Sungai Jempul and

Table 3 Metal evaluation index (MEI) of recreational rivers in Jengka, Malaysia

	WHO ^a					Malaysia ^b				
	Fe	Mn	Pb	MEI	Classification	Fe	Mn	Pb	MEI	Classification
Sg. Jempul	n/a	0.18	17	17.18	medium	0.48	0.70	3.40	4.58	low
Sg. Weh	n/a	0.05	19	19.05	Medium	0.31	0.20	3.80	4.31	Low

n/a Not available, ^aBased on WHO (2011) maximum acceptable concentration, ^bBased on National Drinking Water Quality Standard (NDWQS 2019)

Table 4 The correlation matrix of metal concentrations

	Fe	Mn	Pb
Fe	1	0.930*	-0.730*
Mn		1	-0.577
Pb			1

*Correlation is significant at the level of 0.01 (two-tailed)

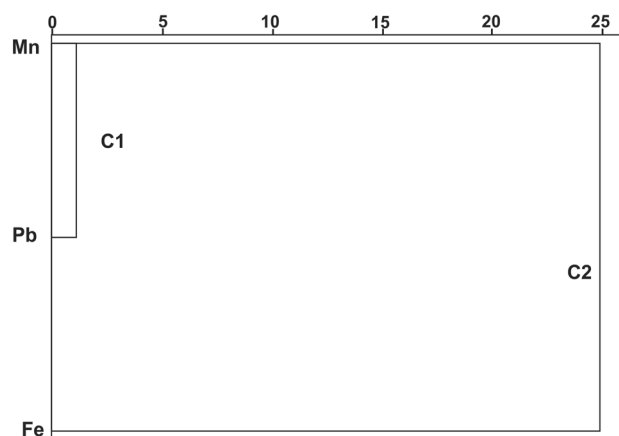


Fig. 2 Hierarchical cluster analysis for Fe, Mn and Pb concentrations from river water

Sungai Weh were found to be medium if based on WHO but low according to NDWQS.

Sources of metal

Table 4 presents the Pearson correlation coefficient between metal concentrations in river water at a confidence level $p < 0.01$. Fe and Mn showed a strong positive relationship ($r = 0.930$). However, Fe and Pb have a negative correlation ($r = -0.730$). A similar negative link between Mn and Pb was observed ($r = -0.577$). Hierarchical cluster analysis for Fe, Mn, and Pb concentrations from river water shows two distinct groups (Fig. 2). Cluster 1 (C1) consists of Mn and Pb and Cluster 2 (C2) comprises Fe, which implies two different potential sources of metal concentrations. Perhaps metal in C1 originates from anthropogenic activities and C2

comes from natural sources. Metals in C1 could be due to significant agricultural activities nearby via surface runoff (Razali et al. 2021; Ghannam 2021; Abdullah et al. 2020; Che Nadzir et al. 2019). Note that both rivers flow through palm oil plantation areas. Lim et al. (2013) suggest that Fe derives from the natural environment.

Human health risk

Table 5 shows the average daily dose (ADD) of Fe, Mn, and Pb from oral and dermal contact in adults and children. In this study, long-term exposure via two pathways was calculated to be 24 and 6 years for adults and children, respectively. Children ADD of metals was reported to be 4.80×10^{-2} mg/kg/day (Sungai Jempul) and 3.42×10^{-2} mg/kg/day (Sungai Weh) for oral exposure to water, which was greater than for adults at 2.29×10^{-2} mg/kg/day (Sungai Jempul) and 1.64×10^{-2} mg/kg/day (Sungai Weh), respectively. Another key route of exposure for the metals investigated is dermal contact with water. Through skin contact with water, hazardous metals can be absorbed in several ways, including swimming, bathing, and washing. Sungai Jempul and Sungai

Weh are recreational rivers. The ADD for dermal contact, however, was shown to be lower than for oral exposure.

The potential non-carcinogenic harmful effects of metals are frequently quantified using the HQ method. When the HQ value surpasses 1 ($HQ > 1$), persons may experience harmful health impacts (non-carcinogenic risk), according to HQ criteria (USEPA 2011). Pb oral exposure by adults and children showed $HQ > 1$ (Table 5). The results, however, contrast to a study by Karim (2011). In general, children have a higher potential non-cancer risk than adults. Children have a smaller body mass than adults and are thus exposed to more metals in terms of body weight (Qu et al. 2012). The cumulative effect of exposed metals is the hazard index (HI). There was a likelihood of non-cancer risk of all metals for adult and child health when evaluating total exposure (HI) via oral and dermal contact. According to Qu et al. (2012), HI is used as a conservative assessment method to compute high-end risk, rather than a low-end risk. Figure 3 shows the HI values from oral and skin contact exposure by adults and children. HI value for children shows higher than the HI value for adults. This finding indicates a chronic non-cancer risk is likely to occur, and the possibility increases with the increase in HI values.

Table 5 Average daily dose (ADD) and hazard quotient (HQ) for adults and children

		Adults				Child			
ADD	River	Fe	Mn	Pb	Total	Fe	Mn	Pb	Total
Dermal	Sg. Jempul	3.05×10^{-9}	4.45×10^{-10}	1.08×10^{-9}	4.58×10^{-9}	1.79×10^{-8}	2.61×10^{-9}	6.34×10^{-9}	2.69×10^{-8}
	Sg. Weh	1.91×10^{-9}	1.40×10^{-10}	1.22×10^{-9}	3.27×10^{-9}	1.12×10^{-8}	8.21×10^{-10}	7.16×10^{-9}	1.92×10^{-8}
Oral	Sg. Jempul	1.53×10^{-2}	2.23×10^{-3}	5.42×10^{-3}	2.29×10^{-2}	3.20×10^{-2}	4.66×10^{-3}	1.13×10^{-2}	4.80×10^{-2}
	Sg. Weh	9.57×10^{-3}	7.02×10^{-4}	6.12×10^{-3}	1.64×10^{-2}	2.00×10^{-2}	1.46×10^{-3}	1.28×10^{-2}	3.42×10^{-2}
HQ	River	Fe	Mn	Pb		Fe	Mn	Pb	
Dermal	Sg. Jempul	5.09×10^{-5}	8.49×10^{-8}	2.06×10^{-7}		2.98×10^{-4}	4.97×10^{-7}	1.21×10^{-6}	
	Sg. Weh	3.18×10^{-5}	2.66×10^{-8}	2.32×10^{-7}		1.86×10^{-4}	1.56×10^{-7}	1.36×10^{-6}	
Oral	Sg. Jempul	2.18×10^{-2}	6.34×10^{-1}	1.03		4.57×10^{-2}	1.32	2.15	
	Sg. Weh	1.36×10^{-2}	1.99×10^{-1}	1.16		2.85×10^{-2}	4.16×10^{-1}	2.43	

Bold values indicate high non-cancer risk

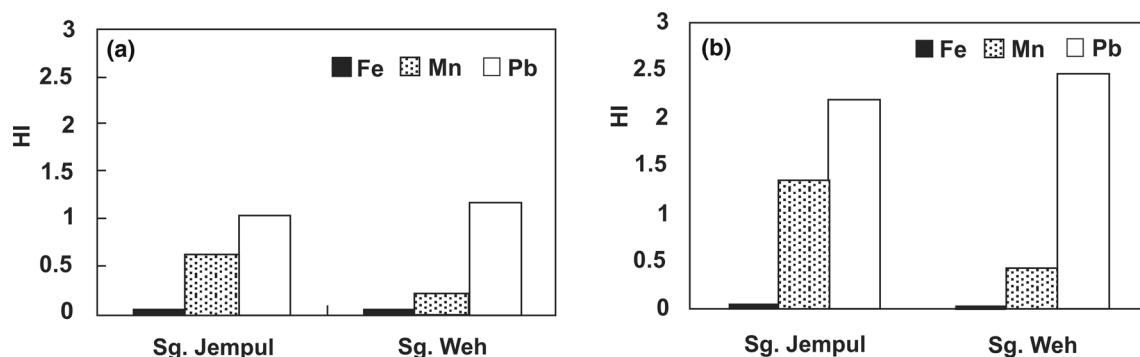
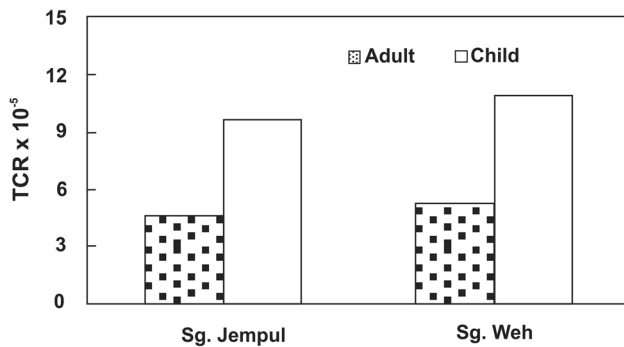


Fig. 3 Hazard index (HI) of metals for **a** adults and **b** children

Table 6 Cancer risk (CR) of Pb for adults and children

		Adults	Child
Dermal	Sg. Jempul	9.20×10^{-12}	5.39×10^{-11}
	Sg. Weh	1.03×10^{-11}	6.09×10^{-11}
Oral	Sg. Jempul	4.61×10^{-5}	9.63×10^{-5}
	Sg. Weh	5.21×10^{-5}	1.08×10^{-4}

Bold value indicates high cancer risk

**Fig. 4** Total cancer risk of Pb exposure from Sungai Weh and Sungai Jempul

The potential of developing cancer during one's lifetime because of exposure to carcinogens is defined as a cancer risk (Li et al. 2014). This risk was estimated for Pb exposure through oral and dermal contact as it is classified as probably carcinogenic to adults and children (ATSDR 2019). The cancer risk (CR) of Pb was assessed for adults and children based on the tolerable range by USEPA (2011). Pb oral exposure for children in this study shows a slightly higher cancer risk than the acceptable limit ($CR = 1.08 \times 10^{-4}$) (Table 6). The risk of cancer from oral exposure is higher than that from skin contact. Based on the total cancer risk value in Fig. 4, children may expose to a cancer risk greater than adults. Similar findings were discovered by Proshad et al. (2020). The total carcinogenic risks from the investigated river water for oral and dermal contact pathways were lower than the limitation value. However, if metal concentrations increase, the anticipated cancer risk in the study area's target population (adults and children) may increase.

Conclusion

This study found that metal in the recreational rivers' water had harmful consequences for the ecosystem and human health. Metal concentrations in recreational rivers followed the trend $Fe > Pb > Mn$. Hierarchical cluster analysis suggested two different groups of metal origin, namely anthropogenic (agricultural) and natural sources. The metal

evaluation index (MEI) values indicated low and medium metal pollution levels. The total heavy metal toxicity load (HMTL) estimated a Pb toxicity in both river water exceeding the allowed limits. Pb poses potential non-cancer to adults and children via the oral pathway. Children are susceptible to the cancer risk of oral Pb exposure ($CR > 10^{-4}$). River water should be free of toxic metals because it is used for a variety of purposes. Comprehensive monitoring of metal elements, such as Cd, Cu, and Zn, and water quality parameters, such as DO, pH, and BOD, is needed. Perhaps a thorough health risk assessment should be dealt with seriously, as the affected population may be exposed to both cancer and non-cancer risks.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s13201-022-01643-1>.

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Author contributions FRS, CMFHCMS and MHAR planned the study. FRS analysed the data and wrote the manuscript. CMFHCMS and MHAR collected the sample and analysed the sample. NS verified, reviewed and edited the manuscript. All authors have approved the final version of the manuscript.

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Data availability Data are available upon request to the corresponding author (fazrul@uitm.edu.my).

Declarations

Conflict of interest The authors declare no conflict of interest.

Ethical approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

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